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FIRE SPREAD THROUGH STRATIFIED FUEL-AIR MIXTURES

A PROJECT FUNDED BY THE
OCCUPATIONAL HEALTH AND SAFETY
HERITAGE GRANT PROGRAM

FIRE SPREAD THROUGH
STRATIFIED FUEL-AIR MIXTURES

UNIVERSITY OF CALGARY
DEPARTMENT OF MECHANICAL ENGINEERING
CALGARY, CANADA

Fire Spread Through Stratified Fuel-Air Mixtures

by

G. A. Karim

A Final Report Describing Research Work Carried Out
Under Agreement No. 85-51R Over the Period April 1986
to September, 1989.
Submitted to Alberta Occupational Health and
Safety Heritage Grant Program.

December, 1989

This is a report that describes the research work that was carried out with the support of Alberta Occupational Health and Safety Heritage Grant Program in relation to the examination of the processes of ignition, flammability and subsequent fire spread within stratified fuel-air mixtures resulting from the convective diffusion of a fuel gas or vapour released into the air at ambient conditions. Predictive procedures were also developed. A number of papers arising from this research are enclosed.

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Abstract

A great number of hazardous situations can be cited arising from the uncontrolled leakage or discharge of a combustible vapour into the immediate surroundings. The research carried out was aimed at establishing a better understanding of the processes of formation of flammable/explosive mixtures and subsequent fire spread in environments wherever fuels are handled, utilized or stored. It was also aimed at providing information for aiding the development of means for the protection against fires and explosions and their subsequent suppression.

The research examined the processes of ignition, flammability and subsequent fire spread within stratified fuel-air mixtures resulting from the convective diffusion of a fuel gas or vapour released into the air at ambient conditions. Predictive procedures were developed and checked against experiment. The analytical part of the investigation involved relatively simple models that predict the concentration changes with respect to time and space in a number of relatively simple configurations following the release of either a fixed mass of a gas or the continuing discharge of a gas following a rupture or a leak into the environment. A number of situations that involved the diffusion of a gas or a gaseous fuel mixture into air or other gaseous media were considered, both without and with the presence of buoyancy effects. The aim was to establish for a number of configurations the arrival and subsequent subsidence of the resulting flammable region at any location.

This was approached by developing a variable effective diffusivity coefficient that is primarily a function of the measured instantaneous fuel concentration gradient and the fuel gas system employed. Further analytical work also considered the common situation of the development of a flammable region following the discharge of a fuel into the atmosphere in the form of a turbulent jet, such as following a leak from a fuel reservoir or a pressurized line. The role of the presence or accumulation of some "background" fuel in the environment was also considered. It was shown for example that the size of the flammable zone formed can be increased very significantly on the presence of some fuel in the surrounding background.

Generalized, relatively simple procedures were also developed for predicting the fuel concentration profiles and their changes with time and space following the release of a fuel into air, typically within a confined space at constant pressure and temperature and for a wide range of fuels. It was based on the one dimensional diffusion equation and an effective eddy diffusivity to be established for the fuel and the situation being considered. Such generalized plots can be employed to

establish the size of the associated flammable zones and their changes with time, including the cases when convective diffusion may be involved.

It was shown that an effective practical way for establishing whether a flammable mixture has been developed at any location is through the provision of an intermittent electric discharge in the form of an electric spark that should produce a propagating flame in the presence of a flammable mixture. This approach was used extensively, yielding information regarding the hazards associated with developing stratified mixtures.

The release of methane upwards into air can generate flammable mixtures at locations above the initial fuel-air interphase extremely rapidly. In the experimental set-up employed this time was a matter of a small fraction of a minute. The diffusion of a heavier fuel gas such as propane, on the other hand, under the same test conditions, takes a very long time to produce similarly a flammable mixture. This representative trend underscores the role of the widely varying diffusional characteristics of the fuel in establishing the flammability characteristics of the stratified mixtures that are formed.

Of particular significance is the discovery of the important phenomenon of the flash fires that can reoccur repeatedly in fuel tanks that are either presumed empty, or near empty, in the presence of an ignition source. It was observed that for the same diffusing mixture, in the presence of a periodic passage of an electric spark, typically every two seconds, a reoccurrence of this flame propagation can take place repeatedly with varying relatively long time intervals between. This indicates that a flammable zone of significant size can be formed between these flame flashovers. This zone results from the subsequent diffusion of some of the remaining unburnt fuel upwards and some fresh air supply downwards, as the hot products of combustion migrate also upwards. Obviously, this reoccurring and delayed fire spread within a stratified mixture that has been repeatedly consumed through flame propagation has a far reaching safety implication in relation to enclosures that could have contained some fuel and had suffered extensive flash fires. Such general trends were also found to varying degrees of severity and frequency with other fuel systems into air within the confines of the long flame tube tested.

On the basis of this research and the mass of available information in the relevant literature, guidelines incorporating safety measures and limits were provided. These were written with the needs of the energy industry in general and the gas industry in particular in mind.

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- Table I ["Some Authored Papers Relating to Safety Research" (May, 1986 - November, 1989).]

- Resulting Publications Cited:

  i) Publication No. 1: "Ignition and Flame Propagation Within Stratified Methane-Air Mixtures Formed by Convective Diffusion" (G. A. Karim and H. T. Lam); Twenty-first Symposium (International) on Combustion/The Combustion Institute, 1986, pp. 1909-1915.

  ii) Publication No. 2: "The Diffusivity of Methane into Air Within Vertical Tubes" (R. Petela and G. A. Karim); Proceedings of the 2nd ASME Fossil Fuel Combustion Symposium, January 14-18, 1989, Tokyo, Japan.

  iii) Publication No. 3: "Examination of the Combustion

of a Fuel Jet in a Homogeneously Premixed Lean Fuel-Air Stream" (G. A. Karim, M. Kibrya, R. Lapucha and I. Wierzba); Proceedings of the International Fuels and Lubricants Meeting and Exposition of the SAE, Portland, Oregon, October 10-13, 1988, Paper No. 881662.

iv) Publication No. 4: "Flame Flashback for Low Reynolds Number Flows" (G. A. Karim and R. Lapucha); published in the AIAA Progress in Astronautics and Aeronautics, Progress Series, (eds. Dr. Allen L. Kuhl et al), Vol. 113, 1988, pp. 367-383.

v) Publication No. 5: "Examination of the Formation of Combustible Mixtures by Diffusional Mass Transfer" (O. A. Badr and G. A. Karim); Transactions of the ASME/Journal of Energy Resources Technology, Vol. No. 111, No. 3, September, 1989, pp. 194-199.

vi) Publication No. 6: "Investigations of the Generation of Flammable Zones Following the Release of a Fuel Gas into the Atmosphere" (G. A. Karim, M. Kibrya, H. Lam, R. Petela and I. Wierzba); Proceedings of the International Specialists Meeting on Major Hazards in the Transport and Storage of Pressure Liquefied Gases, University of New Brunswick. Fire Science Centre, Fredericton, N.B., August 10-13, 1987.

vii) Publication No. 7: "Experimental and Analytical Investigation of the Convective Diffusion of Methane into Air" (G. A. Karim, H. T. Lam, R. Petela and R. Rowe); Transactions of the ASME/Journal of Energy Resources Technology, Vol. 109, No. 4, December 1987, pp. 230-234.

"Some General Guidelines for the Control of Fire and Explosion Within Industrial Establishments" (G. A. Karim).

The research work described was carried out by a number of research personnel, who contributed actively to make the present research possible. The list of personnel involved includes:

R. Petela
C. Y. Zhang
H. T. Lam
O. Badr
M. G. Kibrya
and R. Lapucha

INTRODUCTION

A great number of hazardous situations leading to injury, death, material loss and environmental damage can be cited arising from the uncontrolled leakage or discharge of a potentially hazardous combustible vapour into the immediate surroundings and spreading later on into the environment. The research carried out was aimed at establishing a better understanding of the processes of formation of flammable/explosive mixtures and subsequent fire spread in common environments wherever fuels are handled, utilized or stored. It was also aimed at providing information for aiding the development of efficient means for the protection against fires and explosions and their subsequent suppression.

Overall Objective

The overall objective of the research was to examine the processes of ignition, flammability and subsequent fire spread within stratified fuel-air mixtures resulting from the convective diffusion of a fuel gas or vapour released into the air at ambient conditions. Predictive procedures were to be developed and checked against experiment throughout. Moreover, on the basis of this research and the mass of available information in the relevant literature, guidelines incorporating improved safety measures and limits were to be provided. These were to be written with the needs and experience of the energy industry in general and the gas industry in particular.

Apparatus and Experimental Procedure

Detailed description of the apparatus and experimental procedures employed is provided in the relevant publications enclosed, describing aspects of our work. The phenomena of stratification, ignition and flame propagation were carried out largely within a circular long tube. The concentration gradients are obtained by diffusing the fuel or a retardment to be stratified from a separate chamber at the end of the tube, as shown in Figure (1). The resulting mixture can be ignited electrically at any one of a number of locations after a predetermined time period of interdiffusion. The concentration pattern at any location along the tube can be checked directly using a specially developed transducer based on the change in the velocity of sound within the mixture due to concentration changes along any specified diameter of the tube. Flame propagation can be recorded either by video or a cinecamera. Moreover, occasionally the fibre optical cables installed at intervals along the tube to view the chamber along a diameter were also used. Much of the work carried out involved the use of the buoyant gas, methane, representing natural gas. Other fuels such as propane, ethane, ethylene, carbon monoxide and hydrogen were employed. Retardants such as carbon dioxide, nitrogen and helium were also used.

THE RESEARCH CARRIED OUT

Introduction

The research carried out with the support given by Alberta Occupational Health and Safety Heritage Grant Program was pursued to various degrees of completion and had many objectives and facets. It has yielded numerous contributions to the literature of safety that was published both nationally and internationally. The magnitude of the effort can be judged by the published reports listed in Table I that embody the highlights of our completed contributions. Some of these reports are enclosed. Further aspects of the research need to be continued and pursued to yield very important original contributions to both fire and explosion protection, with particular reference to the energy sector of Western Canada. For example, of particular significance is our discovery of the important phenomenon of the flash fires that can reoccur repeatedly in fuel tanks that are either presumed empty, or near empty, in the presence of an ignition source. This is undoubtedly a complex phenomenon that can be implicated in numerous unexplained mysterious flash fires and explosions involving fuel installations. It requires careful research so as to unravel its complexities and suggest remedies.

Moreover, another interesting and important problem that needs further investigation beyond our initial attempts is the mode of diffusion and associated processes of multi-component homogeneous fuel mixtures into air. Clearly, this is an issue that has a far reaching significance, especially since most gaseous fuels encountered normally involve fuel mixtures. This is a source of uncertainty in relation to safety planning that appears to have been inadequately addressed in the relevant literature.

Analytical Part of the Investigation

The main aim of the analytical part of the investigation was to develop effective yet relatively simple models that can predict the concentration changes both with respect to time and space in a number of relatively simple uniform configurations following the release of either a fixed mass of a gas or the continuing discharge of a gas following a rupture or a leak into the environment. Much progress was made in this regard in a number of publications that were published internationally. A number of situations that involved the diffusion of a gas or a gaseous fuel mixture into air or other gaseous media was considered, both without and with the presence of buoyancy effects. The aim was to establish for a number of configurations the arrival and subsequent subsidence of the resulting flammable region at any location. This is achieved from the prediction of the concentration profile with respect to time and space. In this regard the following cases were considered:

(a) The concentration changes following the release of a buoyant gas such as methane into air both in the open and in a closed system was established experimentally as described in Publication No. 1 (Karim and Lam) using the specially developed velocity of sound method. We came to the conclusion that there is a need to develop a relatively simple approach for the modelling of the diffusion of the highly buoyant gas methane into air that capitalized on the availability of such original concentration data obtained experimentally. Accordingly, the varying concentrations were modelled using such data. This problem was approached by developing a variable effective diffusivity coefficient that is a function of the most important variables influencing the convective diffusion. It was shown that this effective diffusivity can be correlated primarily as a function of the measured instantaneous fuel concentration gradient and the fuel gas system employed. The transient response of the fuel concentration obtained this way was checked throughout experimentally. It was further shown how this approach can be used to predict the development of flammable zones following the release of a fuel gas with time at any location. The approach followed together with the appropriate supporting work and evidence is described in Publication No. 2 (Petela and Karim). This is an ASME paper that is to be part of the forthcoming ASME Energy Conference in New Orleans, January, 1990. Progress has also been made towards establishing the transient conditions for which this modelling can be applied with some confidence. For example, for the case of the release of a buoyant fuel gas, such as methane, into air within a long vertical cylinder, a more complex presently yet unavailable model is needed when considering regions very close to the point of discharge and/or immediately after the release of the gas. Away from these critical conditions, the simplified approach developed appears to be quite effective and adequate.

(b) The release of a fuel mixture made up of two or more components into air was considered both analytically and experimentally. These fuel mixtures were made up typically of methane-propane, very rich methane-air mixtures, methane-ethylene and methane-carbon dioxide. These fuels and their mixtures were chosen, not only because of their practical importance in relation to the question of safety, but also because they afford a good opportunity for testing the effectiveness of any analytical models that may be developed, particularly when two highly dissimilar fuel components with widely different diffusional and combustion characteristics were involved. It appears the diffusional characteristics of fuel mixtures into air is an important problem that has been largely ignored in the relevant literature. The problem is complex yet very important and needs to be addressed adequately. Obviously, only some limited progress has been made by us in this direction within the time available for the current research. It was conclusively evident that in the typical case of the diffusion of initially homogeneous mixtures of methane and propane upwards in air that with the presence of the lighter fuel component the mixture diffuses very rapidly into the air. This rate is very much faster than the heavier component fuel propane. Thus, it tends to form relatively quickly stratified mixtures involving the lighter fuel component into air above the initial interphase boundary. It was shown that should an ignition source be placed somewhere downstream of this initial fuel-air interphase, then a flammable zone may soon develop with time. This zone will be associated mainly with the methane. Once a fuel mixture just heavier than air, that contains less than 52% methane, is reached, then the time taken to produce flammable mixtures will be so long that the heavy fuel component propane will produce mainly the flammable regions. Formulating predictive approaches for the consequences of these complex phenomena is highly desirable. However, this cannot be done reliably. A very approximate approach has been tried with some success. It is based on the assumption that the individual components of the mixture diffuse into air, as well as into each other, in accordance with their corresponding individual estimated effective diffusivity values. The results of this simple approach was checked against some experimental results and was found to yield fairly reasonable agreement.

(c) Further analytical work also considered the commonon situation of the development of a flammable region following the discharge of a fuel into the atmosphere in the form of a turbulent jet, such as following a leak from a fuel reservoir or a pressurized line. The role of the presence or accumulation of some "background" fuel in the environment was also considered. Some of the analytical results obtained were checked against some experimental findings available. It was shown for example that the size of the flammable region arising from the discharge of the fuel in the form of a jet can be established in terms of the initial jet diameter and the corresponding flammability limits for the fuel. The size of the flammable zone formed can be increased very significantly on the presence of some fuel in the surrounding background. This very large growth in the flammable zone on the addition of some fuel with the surrounding air points to the increased explosion and fire hazards associated with the discharge of a fuel from an orifice or a leak in a poorly ventilated room. To base the size and rate of growth of the flammability region on the assumption of homogeneous mixing of the discharging fuel with the room air can severely underestimate the seriousness of the resulting hazards. The results of this aspect of the work are described in Publication No. 3 (by Karim, Kibrya, Wierzba and Lapucha). Moreover, some further relevant information appeared in Publication No. 4 (by Karim and

Lapucha).

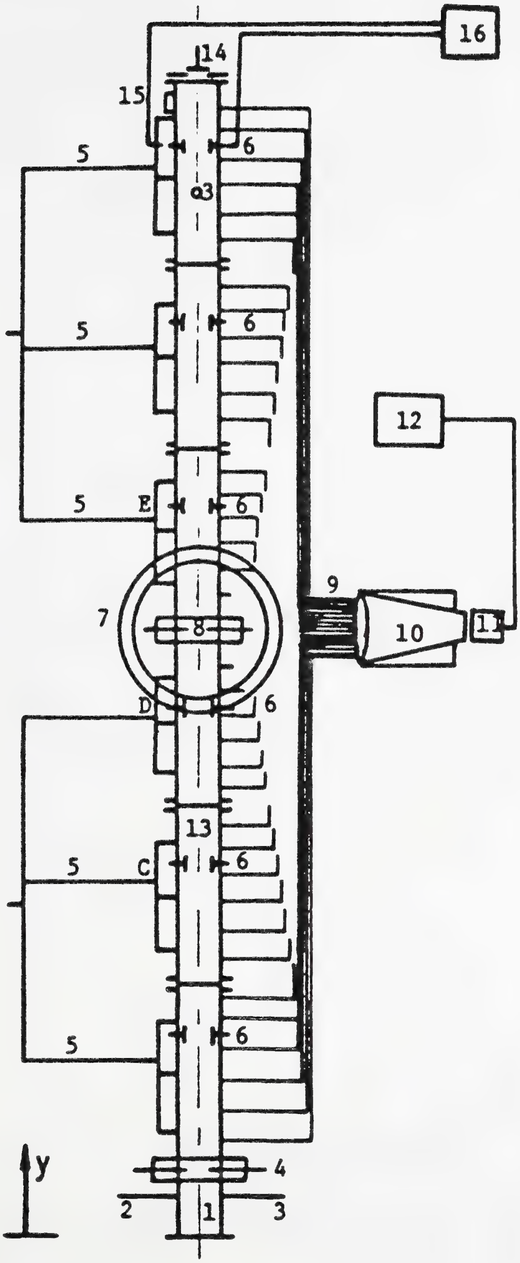
(d) We have developed probably for the first time ever,generalized relatively simple procedures for predicting the fuel concentration profiles and their changes with time and space following the release of a fuel into air, typically within a confined space at constant pressure and temperature. This approach was generalized for a wide range of fuels. It was presented using generalized charts based on the one dimensional diffusion equation and an effective eddy diffusivity to be established for the fuel and the situation being considered. It is suggested that such generalized plots can be employed to establish relatively easily the size of the associated flammable zones and their changes with time, including the cases when convective diffusion may be involved. The approach is described in Publication No. 5 (by Badr and Karim) which includes the presentation of examples involving some typical common fuels diffusing into air.

EXPERIMENTAL CONTRIBUTIONS

(a) It was confirmed that the transient concentration of a diffusing gas into another gas or air can be established very quickly and repeatedly at any location by using the velocity of sound method that was developed earlier by us. Much work was carried out to improve on the accuracy of such measurements, as well as to automate the readings, etc. so as to be able to handle the very large number of observations to be made over a protracted period of time during diffusion. On the basis of the experimental data obtained, a variable effective diffusivity criterion for the upward diffusion of methane was developed and tested. Moreover, it was shown for non-convective molcular diffusion such as the diffusion of methane downwards into air or the diffusion of propane upwards into air that the molecular diffusion theory assumption then was capable of yielding predictions that are in very good agreement with experiment. This agreement was so good generally that the molecular diffusion coefficient for any two gaseous systems can be derived on the basis of the experimental data obtained. However, it became evident that there are also serious limitations with this measuring system, especially when considering the upward diffusion of very buoyant gases such as hydrogen and helium. It is evident that under such conditions the one dimensional diffusion normally assumed for such an analytical system is no longer strictly valid. The rapid diffusion of the buoyant gas upwards into a heavier gas within a tube or cylinder would result in a faster core of moving gas relative to its outer regions. Thus, the assumption of a one dimensional diffusion needed for the proper working of the analyser will no longer be valid. Accordingly, attempts were made to correct analytically for this effect by using a two-dimensional approach. This, however, proved to be very unwieldy.

(b) It was shown that an effective practical way for establishing whether a flammable mixture has been developed at any location is through the provision of an intermittent electric discharge in the form of an electric spark that should produce a propagating flame in the presence of a flammable mixture. This approach was used extensively, yielding very valuable information regarding the hazards associated with developing stratified mixtures. Some aspects of these findings were reported in Publication No. 6 (by Karim, Kibrya, Lam, Petela and Wierzba). It was shown that flame propagation within a stratified mixture arising from an electric spark can proceed at concentrations lower than the established flammability limit under the same conditions had there been a homogeneous mixture. This departure from the concentration of the flammability limit tends to increase the greater the concentration gradient of the fuel in the stratified mixture. Moreover, the ensuing flame propagation tends to be somewhat faster than for the corresponding homogeneous mixture case. Obviously, this has an important implication as far as safety planning is concerned. Moreover, it was shown that there is a need for a stricter definition and control of the energy of the intermittent spark being used to monitor for the first development of a propagating flame. More energetic sparks can ignite mixtures that may appear to be non-flammable. Much effort was expended in the measuring and control of such ignition energy for the tests conducted.

(c) It was evident throughout that the release of methane upwards into air can generate flammable mixtures at locations above the initial fuel-air interphase extremely rapidly. In the experimental set-up employed and described in Figure (1) this time was a matter of a small fraction of a minute. On the other hand, the release of methane into configurations where it is made to diffuse downwards into air, the spread of methane proceeds then by molecular diffusion and takes in comparison an exceedingly long time to generate a flammable zone, as described in Publication No. 7 (by Karim, Petela, Kibrya, and Wierzba). The highly diffusive nature of methane in the upward direction due to its buoyancy is a marked contrast to its sluggishness to disperse downwards. As can be seen in Figure (2), the diffusion of methane upwards into air within the open flame tube of Figure (1) generates a flammable mixture in a matter of seconds at a test location some 340mm above the initial fuel-air interface. The diffusion of propane on the other hand, under the same test conditions, takes a very long time (around 20 minutes) to produce similarly a flammable mixture. As can be seen in Figure (2), the upwards diffusion into air of initially homogeneous mixtures of methane and propane that contain appreciable concentrations of methane is essentially similar to that encountered with methane. However, the upward diffusion into air of initially homogeneous fuel mixtures containing appreciable concentrations of propane, takes even longer time than for pure propane to produce a flammable mixture. This representative trend underscores the role of the widely varying diffusional characteristics of the fuel mixture in



1. FUEL SECTION (LOWER COMPARTMENT)
2. FUEL INLET
3. EVACUATION LINE
4. PLATE VALVE/SHUTTER
5. AIR INLET MANIFOLDS
6. TRANSDUCER PORT/ MOVABLE IGNITION SOURCE
7. ROTATING (INCLINATION) DISC
8. SUPPORTING PLATE
9. FIBRE OPTICS
10. OPTICAL SYSTEM FOR LIGHT CONCENTRATION
11. PHOTO DIODE
12. PHOTO DIODE OUTPUT RECORDER
13. AIR SECTION (UPPER COMPARTMENT)
14. NOZZLE VALVE
15. RUPTURE DIAPHRAGM RELIEF VALVE
16. IGNITION CIRCUIT

Figure 1. Schematic Diagram of the Apparatus Used

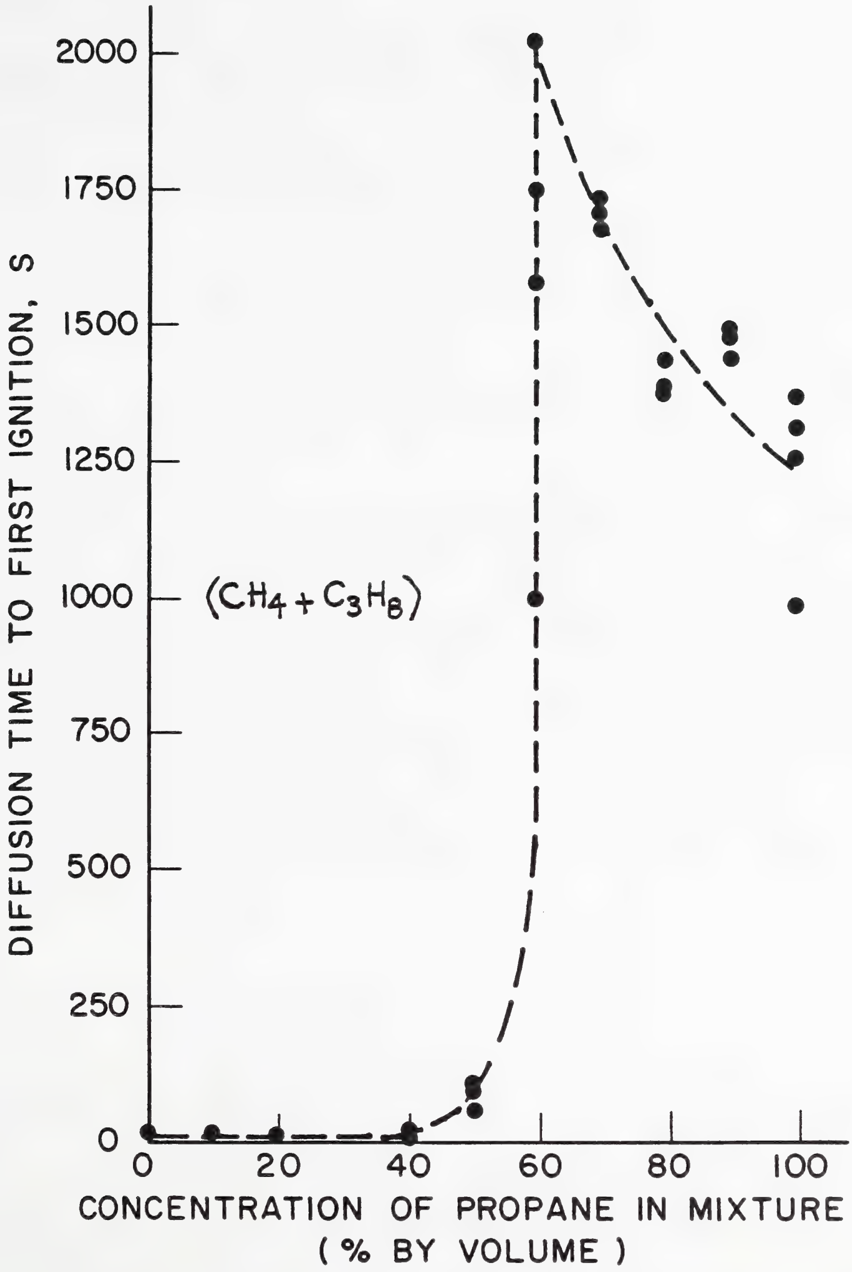


Figure 2. Variations of the Diffusion Time Required to Produce First Successful Flame Propagation Within Stratified Mixtures in the Presence of a Periodic Spark (2 seconds interval) at a Fixed Location of 0.340m from the Fuel-Air Interface, Various Propane-Methane Initial Fuel Mixtures are Involved

establishing the flammability characteristics of the stratified mixtures that are formed. This is also supported by the observation of the behaviour of the diffusional processes of homogeneous mixtures of methane and carbon dioxide in air, shown in Figure (3). Carbon dioxide tends to have somewhat similar diffusional characteristics to propane. It can be seen that the failure to produce a flame at the test location tends to occur again at concentrations of around 50% carbon dioxide in the methane. This is consistent with conditions when the mixture ceases to become buoyant. There are also initial concentrations where it would be impossible to provide sufficient amount of methane at the test section to initiate flame propagation following ignition. This trend is seen with the diffusion of initially homogeneously mixed methane and air upwards into air ($\approx$ 30% CH_4). However, because of the lower density of air in comparison to that of carbon dioxide and the relative superior flammability of methane-air mixture in air in comparison to that for methane-carbon dioxide in air, the initial concentrations of methane that just fail to produce flammable mixtures tends to be slightly higher.

d) As has been described earlier, procedures have been developed to check at any location along the vertical flame tube, the minimum diffusion time needed for the formation of a flammable mixture. This was achieved through the passage of a periodic spark at the location and noting the time needed for a propagating flame to be initiated within the mixture by the spark. Due to the stratified nature of the mixture, such a fire spread will tend not to engulf fully the whole length of the tube. A mixture too rich to support combustion may still be present at

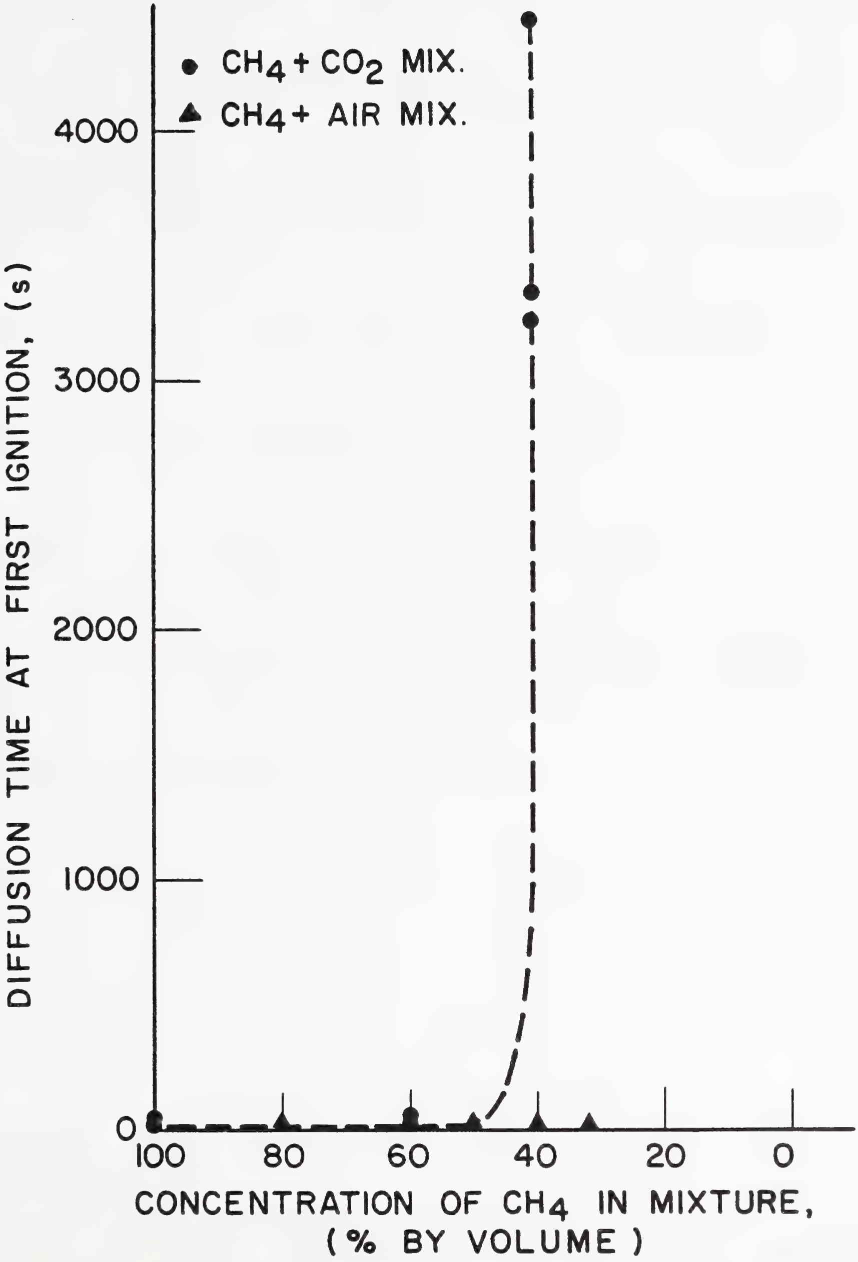


Figure 3. Variations of the Diffusion Time Required to Produce First Successful Flame Propagation Within Stratified Mixtures in the Presence of a Periodic Spark (2 seconds interval) at a Fixed Location of 0.340m from the Fuel-Air Interface; Various Methane-Carbon Dioxide and Methane-Air Initial Fuel Mixtures are Involved

the lowest part of the tube, while a mixture much too lean to support combustion may exist at its top. Expectedly, the buoyant fuel methane diffuses upwards so quickly that a flame would propagate in a matter of seconds after initiating diffusion at a typical location of 0.34m above the initial fuel-air interface. This is, as indicated earlier, compared to the relatively much longer diffusion times needed for flame propagation, typically 16-23 minutes for the corresponding cases involving the upward diffusion of ethane or propane into air. It was observed that for the same diffusing mixture, in the presence of a periodic passage of an electric spark, typically every two seconds, a reoccurrence of this flame propagation can take place repeatedly with varying relatively long time intervals between. This is shown typically in Figure (4) for the case of the diffusion of propane into air. This indicates that a flammable zone of significant size can be formed between these flame flashovers. This zone results from the subsequent diffusion of some of the remaining unburnt fuel upwards and some fresh air supply downwards, as the hot products of combustion migrate also upwards. Such a hypothesis was tested by comparing the calculated time required to generate a flammable mixture at the spark location following a flame propagation. This is done on the basis that the remaining mixture that was too rich to burn diffused upwards into air between flame flashovers to generate once more a flammable mixture. The rest of the fuel associated with initially formed lean mixtures would have been consumed by the flame propagation. This approach was considered repeatedly following every fire flashover. As shown typically in Figure (5), this approach produced good agreement with experiment for the first few flame flashovers. However, very large deviations between experiment

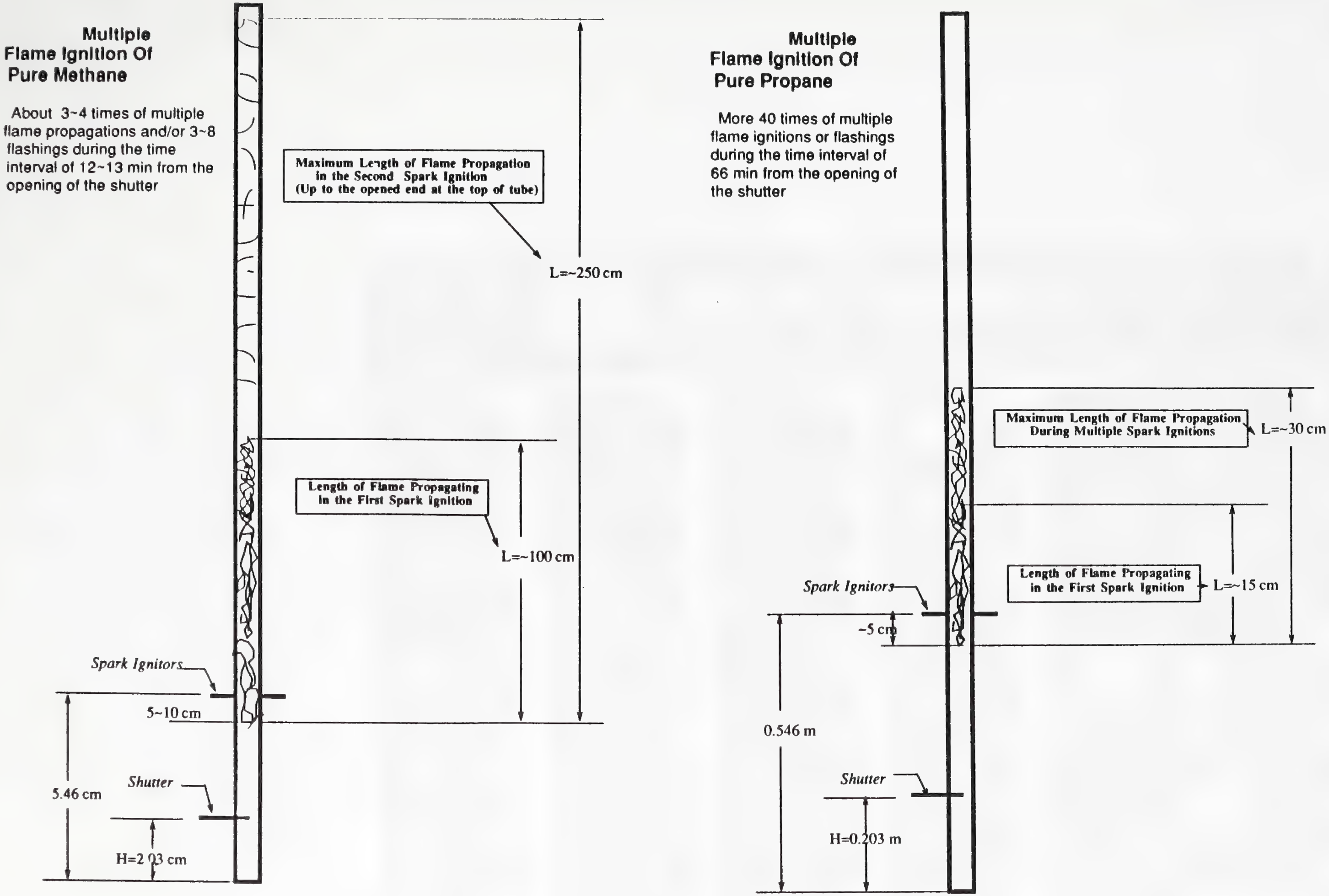

Multiple Flame Ignition Of Pure Methane
About 3~4 times of multiple flame propagations and/or 3~8 flashings during the time interval of 12~13 min from the opening of the shutter
Maximum Length of Flame Propagation in the Second Spark Ignition (Up to the opened end at the top of tube)
L=~250 cm
Length of Flame Propagating in the First Spark Ignition
L=~100 cm
Spark Ignitors
5~10 cm
5.46 cm
Shutter
H=2 03 cm
Multiple Flame Ignition Of Pure Propane
More 40 times of multiple flame ignitions or flashings during the time interval of 66 min from the opening of the shutter
Maximum Length of Flame Propagation During Multiple Spark Ignitions
L=~30 cm
Length of Flame Propagating in the First Spark Ignition
L=~15 cm
Spark Ignitors
~5 cm
0.546 m
Shutter
H=0.203 m

Figure 4a. Schematic Representation of Typical Multiple Flame Propagation in the Set-Up Employed Involving Methane Diffusion into Air and Propane Diffusion into Air

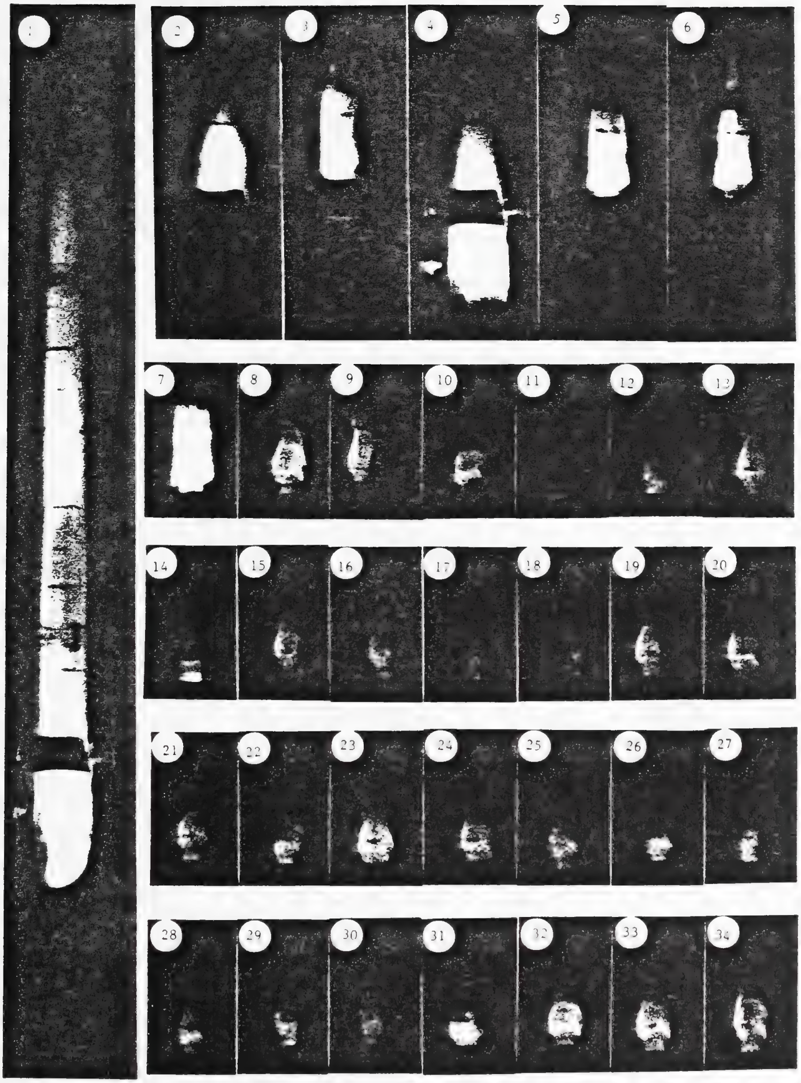


Figure 4b. Typical Photographic sequence Showing the Multiple Flame Flashes in a 60% Methane - 40% CO_2 Mixture Diffusing Upwards into Air in the Presence of a Periodic Spark

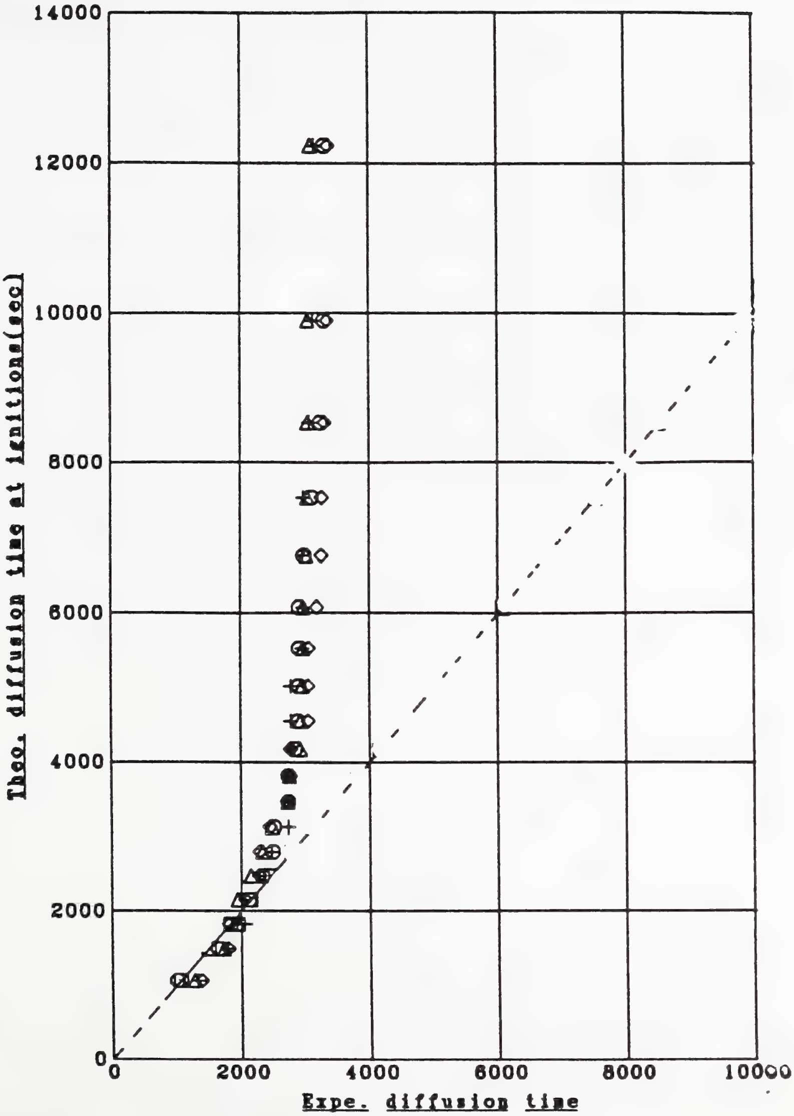


Figure 5. Comparison Between the Experimental Time Observed for the Sequence of Multiple Flame Flashes with Predicted Time for the Diffusion of Pure Propane Upwards into Air at a Location 0.340m Above the Initial Fuel-Air Interface

and predictions developed subsequently. Undoubtedly this is due largely to the very complex physical and chemical processes involved and the simplistic diffusional model employed that ignores the roles of heating and contamination of the mixtures with combustion products as a result of the repeated flame propagation.

Obviously, this reoccurring and delayed fire spread within a stratified mixture that has been repeatedly consumed through flame propagation has a far reaching safety implication in relation to enclosures that could have contained some fuel and had suffered extensive flash fires. Such general trends were also found to varying degrees of severity and frequency with other fuel systems into air within the confines of the long flame tube tested.

Concluding Remarks

The research carried out with the support given by Alberta Occupational Health and Safety Heritage Grant Program was pursued to various degrees of completion and had many objectives and facets. It has yielded numerous important contributions to the knowledge of the field of safety and fire and explosion hazards. It is evident that this research needs to be continued and pursued to yield very important original contributions to both fire and explosion protection, with particular reference to the energy sector of Western Canada. Research in this area is badly needed more than ever before.

TABLE I

Some Authored Papers Relating to Safety Research

May, 1986 - November, 1989

TABLE I

Some Authored Papers Relating to Safety Research

May, 1986 - November, 1989

The following list of papers were all authored by Dr. G. A. Karim, Department of Mechanical Engineering, The University of Calgary, Calgary, Alberta, Canada T2N 1N4 (or co-authored as indicated in parentheses below):

"The Rich Flammability Limits of Methane-Gaseous Fuels-Diluents Mixtures," (co-author I. Wierzba). Proc. 1989 International Gas Research Conference, November 6-9, 1989, Tokyo, Japan.

"The Diffusivity of Methane into Air Within Vertical Tubes," (co-author R. Petela). Proc. 2nd ASME Fossil Fuel Combustion Symposium, January 14-18, 1990, New Orleans, Louisiana.

"A Predictive Approach for the Flammability Limits of Methane-Nitrogen Mixtures," (co-author I. Wierzba). Proc. 2nd ASME Fossil Fuel Combustion Symposium, January 14-18, 1990, New Orleans, Louisiana.

"Examination of the Multiple Reoccurrence of Fire Spread Within a Stratified Mixture in the Presence of an Ignition Source," (co-author C. Y. Zhang), Proc. 1989 Spring Technical Meeting, The Combustion Institute, Canadian Section, Toronto, Ontario, May 31-June 2, 1989.

"Flame Initiation and Spread Characteristics Within Homogeneous Streams of Methane-Air Mixtures," (co-author G. Sarpal), Transactions of the ASME/Journal of Energy Resources Technology, Vol. 111, September, 1989, Number 3, pp. 200-204.

"Examination of the Formation of Combustible Mixtures by Diffusional Mass Transfer," (co-author O. A. Badr), Transactions of the ASME/Journal of Energy Resources Technology, Vol. No. 111, Number 3, September, 1989, pp. 194-199.

"The Flammability of Fuel Mixtures in Air Containing Propane and Burane," (co-author I. Wierzba), Transactions of the ASME/Journal of Energy Resources Technology, Vol. No. 111, Number 2, June 1989, pp. 100-103.

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PUBLICATION NO. 1

"Ignition and Flame Propagation Within Stratified Methane-Air Mixtures Formed by Convective Diffusion" (G. A. Karim and H. T. Lam); Twenty-first Symposium (International) on Combustion /The Combustion Institute, 1986, pp. 1909-1915.

Twenty-first Symposium (International) on Combustion/The Combustion Institute, 1986 pp. 1909–1915

IGNITION AND FLAME PROPAGATION WITHIN STRATIFIED METHANE-AIR MIXTURES FORMED BY CONVECTIVE DIFFUSION

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The concentration changes with time and space of methane following its release into air, under the combined effects of molecular diffusion and natural convection were examined. A smooth, cylindrical, vertical flame tube of 116.0 inches (2.946 m) long and 2.5 inches (63.5 mm) inside diameter was used. The tube was separated into two sections by a plane shutter: a lower small fuel section and an upper large air section. Fuel stratification, caused mostly by convective diffusion, was initiated following the opening of the shutter. The concentration profile as a function of diffusion time at specified regions of the tube was determined by measuring the velocity of sound in the medium using a specially developed "ultra-sonic gas analyzer". To predict the experimental concentration profile in such a system an eddy diffusivity that is a function of both the diffusion time and the local concentration gradient is needed.

The range of diffusion time within which a flammable zone was developed within the stratified mixtures was determined at various locations employing a periodic spark source, for downward flame propagation. It was observed that the flammability limits of downward flame propagation within stratified methane-air mixtures were wider than those obtained for the corresponding homogeneous quiescent mixtures.

Flame speeds of both accelerating and decelerating flames within stratified methane-air mixtures caused by the natural convection process were found to be higher than those encountered under homogeneous conditions at the corresponding methane concentration. Some possible reasons for the enhanced flame speeds are discussed.

Introduction

Non-homogeneous combustible atmospheres are frequently encountered and these are important, particularly as far as safety is concerned. A great number of actual hazardous situations can be cited that result from the leakage of fuel off storage tanks or pipelines, oil or gas spills during transportation, the formation of gas pockets in coal mine galleries or in overheated nuclear reactors. These situations involve the stratification of a combustible gas and flame propagation within layered atmospheres. A better understanding of the fire spread characteristics within stratified media and the associated mass transfer mechanism for the formation of these mixtures is needed to reduce ultimately the incidence of the many hazardous situations that are frequently encountered.

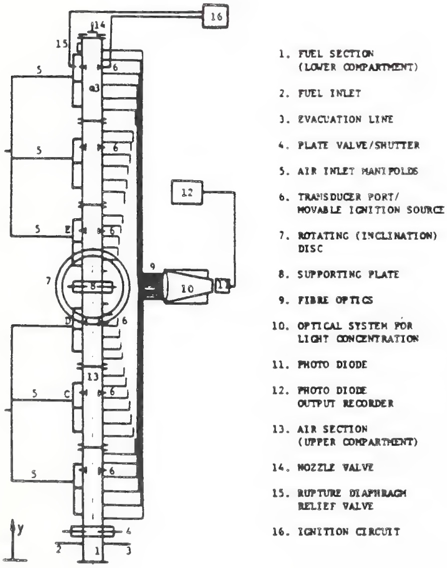
Examination of the relevant literature reveals that essentially very limited amount of research work has been carried out about the flame characteristics within stratified mixtures. Coward and Georgeson[1] studied the movement of methane layers caused by molecular and turbulent diffusion in relation to fire hazards in underground coal mines. Bakke and Leach[2] examined the formation and dispersion of methane roof layers in coal mines due to buoyancy effects. Perlee et al[3] examined the upward flame propagation in the direction of increasing concentration within methane-air strata in vertical tubes. Liebman et al[4] studied the characteristics of flames propagating in vertical tubes both in upward and downward directions through layered methane-air mixtures formed by molecular diffusion of a rich homogeneous dry methane-air mixture into dry air. Tsang[5,6] considered upward flame propagation within stratified mixtures of methane and air obtained by natural convection of two initially different homogeneous mixtures having different densities. Badr[7,8] considered also the nature of flame propagation within various stratified mixtures of methane in air both involving increasing or decreasing concentration gradients. Panlilio[9] also observed the behaviour of flames propagating within stratified diluent-fuel-air mixtures.

The present work set out to investigate the formation of a stratified mixture following a release of methane into air at constant pressure, under the combined effects of molecular diffusion and natural convection within a long cylindrical tube. The ignition and flammability characteristics of the resulting mixture were also investigated. Spark ignition and downward flame propagation were employed at ambient pressure and temperature.

Experimental Apparatus

A long circular smooth flame tube of two and half inches (63.5 mm) inside diameter consisting of a nine-foot (2.75 m) upper air section and an eight-inch (0.203 m) long fuel section was employed. The large length to diameter ratio was employed so as to approximate to a one-dimensional situation. The tube assembly and associated apparatus are shown schematically in Fig. 1.

A quick action thin flat plate shutter was connected between the two sections of the tube containing the air and methane. Upon rapid opening of the shutter, natural convection and molecular diffusion allowed both gases to interdiffuse within each other generating in time a stratified atmosphere. The concentrations at any point within the flame tube changed with time. An electric spark of adequate and constant energy was employed to initiate, after a certain specified diffusion time, flame propagation. The spark electrodes could be installed at any of a number of stations along the tube. Moreover, to establish the shortest time needed for the formation of an ignitable mixture at any specific location, intermittent sparking was allowed at a one second interval in that location to test whether a flame can be made to propagate from that location. Similarly, the longest diffusion time that can allow a flame propagation at the same location was also determined. Accordingly, the minimum and maximum diffusion times permitting a downward propagating and self-sustaining flame at any location were established experimentally with the periodic spark source. Usually sparking was initiated just prior to the reaching of these limits. However, it was found that earlier sparking did not affect the results to any detectable extent.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1. FUEL SECTION (LOWER COMPARTMENT)
2. FUEL INLET
3. EVACUATION LINE
4. PLATE VALVE/SHUTTER
5. AIR INLET MANIFOLDS
6. TRANSDUCER PORT/ MOVABLE IGNITION SOURCE
7. ROTATING (INCLINATION) DISC
8. SUPPORTING PLATE
9. FIBRE OPTICS
10. OPTICAL SYSTEM FOR LIGHT CONCENTRATION
11. PHOTO DIODE
12. PHOTO DIODE OUTPUT RECORDER
13. AIR SECTION (UPPER COMPARTMENT)
14. NOZZLE VALVE
15. RUPTURE DIAPHRAGM RELIEF VALVE
16. IGNITION CIRCUIT

Fig. 1. Schematic Diagram of the Apparatus

A spark gap of around 6.3 mm and stainless steel cylindrical electrodes of 1.5 mm diameter were employed. The form of the energy release during the spark discharge was examined by recording dynamically on an oscilloscope the voltage and current traces across the horizontal spark gap. The total energy of the spark discharge was then determined by graphical integration and found to be around 90 mJ.

An approach involving the use of fiber optical cables was developed to determine the apparent speed of the propagating flame within the tube. The cables were placed normally along the length of the upper tube to view the inner tube walls along a diameter. The light energy emitted by the propagating flame front as it arrives at the location of the cable end was transmitted by the cable to a photomultiplier whose electrical output was displayed with respect to time on a strip chart recorder. A record of the change of the output of the photomultiplier seeing the ends of a bundle of flame viewing cables with respect to time allowed the determination of the instantaneous flame propagation rate from the times of the arrival of the flame front at the stations of the various light cables. The position of the flame front within the tube was also checked by video recording.

To determine experimentally the instantaneous average gas concentration along a tube diameter at different test locations a "sonic gas analyzer" of our design was employed. The velocity of sound in a gas mixture is a function of its density. Therefore, through the passage of an ultrasonic pulse across a tube diameter at the test location, it is possible to determine the concentration of the species in a binary mixture of gases from the measurement of velocity of sound in the mixture. Changes in the concentrations due to diffusion with time can thus be established. A calibration curve was obtained by measuring the velocity of sound, for accurately known and prepared homogeneous methane-air mixtures within the flame tube under ambient pressure and temperature. This experimentally derived curve was used subsequently for determining the changes in the concentration with time within the tube. The mean time interval for the ultrasonic pulse (40 kHz) to travel across the tube was obtained by recording fifty readings using a data acquisition system. The associated error was assessed to be ± 0.08%.

The conditions considered in this work involved the stratification of methane in air through the rapid opening of a full-area, thin and smooth plate valve. The stratification of methane in air in the radial direction, though it may exist, was assumed to be small compared to the axial gradients away from the initial interphase region. This simplifying assumption was partly supported by tracer smoke visualization studies and by the observation that at various axial locations during the ignition tests, the initial downward flame front appeared to be relatively flat. It is acknowledged that if the methane was released from a point or a line source into air simulating a leak then this view may need to be modified. Moreover, similar experiments involving the release of the more buoyant helium into air indicated that such a one-dimensional assumption may be unjustified. No other similar tests were carried out in apparatus having a substantially different size or geometry from the one employed in this investigation.

Changes in the Concentration of Methane with Time due to Convective Diffusion

The concentration of methane in air as a function of diffusion time at a number of locations along the flame tube was established using the ultrasonic gas analyzer. Figure 2 shows typical experimental results for the conditions, both when the tube was closed and when it had an open upper end, at three locations, 'C', 'D' and 'E', at distances of 30.0" (0.762 m), 48.0" (1.219 m) and 66.0" (1.676 m) respectively, away from the plate valve position separating the two sections. Each individual curve though displayed in general a similar trend to that expected from the solution of the one-dimensional molecular diffusion problem with an open end condition, the actual diffusion time observed is drastically faster even when account is made for the role of buoyancy.

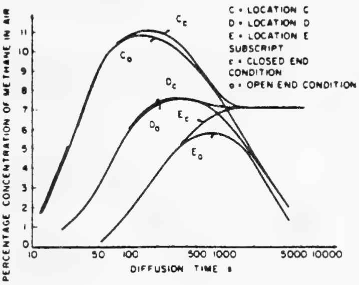

FIG. 2. Comparison of concentration profiles obtained at various locations for the binary system of methane diffusing upwards into air with open and closed upper end conditions—Methane section of 8" (0.203 m) long. The three locations 'C', 'D' and 'E' are at distances 30.0" (0.762 m), 48.0" (1.219 m) and 66.0" (1.676 m) respectively, away from the separation plate valve.

To predict the experimental concentration profile, the molecular diffusion equation needs to be modified such as using an effective eddy diffusivity that can be typically around four orders of magnitude higher than the corresponding molecular diffusivity[10]. An attempt was made to reproduce the concentration profile as a function of diffusion time at any location by solving a modified diffusion equation and using different values for the constant eddy diffusivity. It was evident that the results obtained with a constant value of the eddy diffusivity could not fit the experimental curve well, indicating that such an eddy diffusivity is a function of both diffusion time and concentrations. A very high value is needed for relatively small diffusion times when high concentration gradients exist. As the diffusion time increases lower values are required.

Flammability Characteristics

Experiments were undertaken to determine the time range required at various locations along the tube for a flammable mixture to exist following the release of methane into the air at constant pressure by opening suddenly the shutter separating methane at the lower compartment from the air at the upper compartment of the tube and opening the remote end of the tube to the atmosphere. This time period commences with the minimum diffusion time from shutter opening required for first generating a mixture that would allow a flame to

TABLE I
Summary of Results of Flammability Characteristics of Downward Flame Propagation through Stratified Mixtures of Methane and Air for Both Open and Closed Ends at Various Spark Locations

Boundary Condition	Spark Location	Minimum Diffusion Time (s)	Corresponding Methane Conc. at Initiation of Flame Kernel (%)	Corresponding Conc. Gradient at Initiation of Flame Kernel (% methane/m)	Distance Travelled by Downward Flame from Spark to Extinction (m)	Corresponding Methane Conc. at Flame Extinction (%)
OPEN END	C (y = 0.965m)	24.3	5.05	13.39	0.345	13.80
	D (y = 1.422m)	76.5	5.10	10.24	0.665	13.80
	E (y = 1.880m)	510.0	5.70	3.54	1.880	12.90
CLOSED END	C (y = 0.965m)	23.9	5.05	13.00	0.353	13.85
	D (y = 1.422m)	77.0	5.10	10.24	0.681	13.85
	E (y = 1.880m)	430.8	5.70	3.94	1.880	13.60

propagate at the spark location and ends with the corresponding diffusion time beyond which a flame just fails to propagate at that location. The boundaries of the resulting flammable zone were also determined for different spark locations and diffusion times by establishing the point where the propagating flame front just became extinguished by meeting either a too rich or too lean a mixture.

The experimentally established minimum diffusion time for flame propagation for different spark locations permits the local minimum methane concentration to be read off from the appropriate experimentally determined concentration profile. Table I lists some of the results obtained. It can be seen that the concentration at the minimum diffusion time is relatively lower than the corresponding lean flammability limit of 5.9% by volume, determined for downward flame propagation in the same experimental set-up for homogeneous quiescent mixtures of methane and air. This trend applies to other spark locations, both for open and closed end situations.

The apparent limiting mixtures established under convective diffusion conditions involve neither a homogeneous mixture nor a quiescent state, since vigorous convection took place within the tube. This convective diffusion, particularly just after the commencement of the diffusional process can involve small scale turbulence influencing the local state and concentration of the methane at the spark location. Hence a lower concentration of fuel is needed to permit combustion. The severity of the concentration gradient of the fuel in air at the spark location contributes also to the lowering of the limiting methane concentration at the minimum diffusion time at which a flame could be generated. When determining the flammability limit for a homogeneous quiescent mixture, the resulting flame is required to propagate the whole length of the test tube to consider the mixture flammable. However, in stratified mixtures, the resulting flame propagates shorter distances merely to the location of the limit mixture. Hence, a relatively shorter flammable zone may be generated around the spark location with a concentration lower than the lean limit for homogeneous mixtures. In the presence of a positive concentration gradient at the spark location, the flame front moves to regions of higher methane concentrations releasing enough energy to sustain and even accelerate the flame in the case of lean mixtures.

The transient fuel-air concentration at the igniter location may be outside the lean flammability limit at the time when a successful ignition event was attempted. The region of influence of the igniter may extend beyond this point. Where this region of influence is sufficiently large such as due to strong igniter pulse or steep concentration gradient, downward flame propagation into the regime of richer stoichiometries may occur. In this investigation a spark energy of around 90 mJ and a horizontal gap of 6.3 mm were employed throughout. Tests at locations involving severe concentration gradients were also avoided.

Locations closer to the shutter, and the initial interphase, had smaller values of the minimum diffusion time and higher concentration gradients. This, as shown in Table I, a lower local concentration is needed to initiate a flame at a location near the shutter than at locations away from it. Similarly, the corresponding values of the concentration of the methane in air at the locations where the flame appeared to die out, when predicted from data such as those of Fig. 2, were somewhat higher than the corresponding value of the rich flammability limit when established for homogeneous quiescent mixtures for downward flame propagation. The induced small scale turbulence due to the local concentration gradient contributed to this slightly higher rich limit observed. Moreover, as the flame approached the extinction level, its luminosity decreased rapidly making it difficult to establish with certainty the exact location of flame extinction. Consequently, the concentration at the location where the flame was actually extinguished was probably even higher than the values measured.

It can be noted that for flames initiated at location C, which is relatively near the shutter, the range of the flammable zone appeared to be quite short. The flame travelled downwards merely around 0.330 m before it was extinguished due to over rich mixtures being encountered. For the spark location D, located further away from the initial interphase, the flame travelled 0.660 m before being extinguished. Flame travel all the way down the tube (1.880 m) was observed when ignition occurred at location E. For spark location C, the minimum diffusion time involved was too short to allow the methane to diffuse far enough so that only a short flammable zone was possible. For spark location D, the minimum diffusion time was relatively longer allowing the methane to develop a thicker flammable zone. For spark location E, the minimum diffusion time was sufficiently long allowing the methane to diffuse further and form a flammable mixture all the way down to the bottom of the tube. Figure 3 shows the location of the flammable front with time both when the flame tube was open to the atmosphere and when it was closed. The variation of the apparent lean limit for the methane-air stratified mixture with the local concentration grandient is shown in Fig. 4. As would be expected the observed values are the same whether the tube end was open or closed.

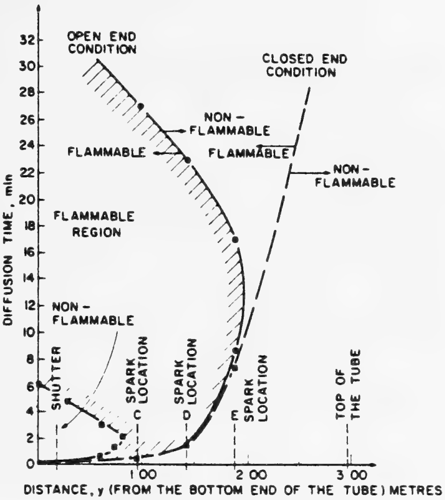

Fig. 3. Flammable regions in closed and open systems following convective diffusion of methane diffusing upwards into air—Methane section of 8" (0.203 m) long and the whole tube of 116" (2.946 m) long.

The Apparent Flame Speed within Stratified Methane-Air Mixtures

The apparent flame propagation rate within the stratified methane-air mixtures were examined. At the minimum diffusion time for

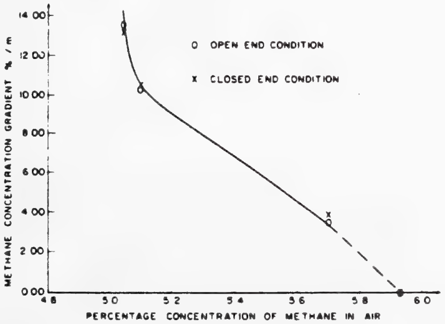

Fig. 4. Relation of the measured methane concentration gradient and the apparent lean limit for downward flame propagation within stratified mixtures.

generating an 'ignitable' mixture at any specified location, a flame was initiated with sparking periodically the mixture at that location. The resulting apparent flame speed values are shown in Fig. 5 as a function of the corresponding local methane concentration. It can be seen that results involving stratified mixtures have the same general trend as those for homogeneous mixtures. However, the apparent flame speeds for both accelerating and decelerating flames within the stratified mixtures tended to be higher than those encountered under homogeneous conditions for the same methane concentration.

Within the lean region, a possible reason for the observed difference in the apparent flame speeds is the convective effect of the increasing methane concentration in the direction of flame propagation. Hence, the flame front will experience a higher concentration than expected and propagate at a higher speed. For flame propagation from the lean limit to the stoichiometric which results in an accelerating flame, pressure waves formed at the flame front and reflected from both ends of the flame tube will interact with the flame front and will tend to create disturbances to the flame front. The resulting changes in the surface area of the flame front would produce corresponding changes in the volumetric rate of burning and thus accelerate the flame towards the unburned gases.

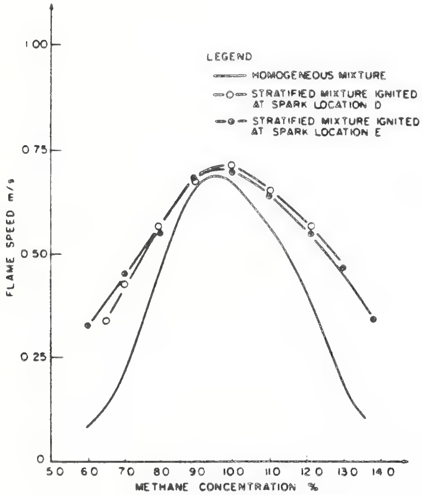

FIG. 5. Comparison of various flame speed profiles obtained from homogeneous methane-air mixture and non-homogeneous stratified methane-air mixtures—downward flame propagation and open upper end condition when ignited at locations 'D' and 'E' at distances 48.0" (1.219 m) and 66.0" (1.676 m) respectively, away from the separation plate valve.

The dominant factor for the enhanced flame speed observed within the rich region is the corresponding change in the amount of heat generated at the flame front and the temperature of the burned gases. For homogeneous mixtures, the amount of energy released at the flame front and the flame temperature remain essentially constant. However, for propagating flames with decreasing velocity from the stoichiometric mixture to the rich limit the rate of heat transfer to the unburnt mixture would be increased through relatively greater conduction and radiation heat transfer due to the higher temperature difference. The flame temperature would be higher for stoichiometric than for rich mixtures and the enhanced preheating of the unburnt gases, compared to the homogeneous conditions, would result in faster local flame speeds compared to those under homogeneous conditions.

Conclusions

For the system involving convective diffusion of methane into air within a long vertical cylindrical tube, the concentration profile observed at any instant could only be made to correspond to that obtained by solving the diffusion equation when using varying values of an "eddy diffusivity" that can be orders of magnitude higher than the corresponding molecular diffusivity.

The limits for downward flame propagation determined within stratified methane-air mixtures appeared to be wider than the corresponding limits obtained for homogeneous quiescent mixtures. For stratified mixtures, the concentration gradient at the spark location appeared to affect the lean limit. As the gradient increased, the lean limit tended to decrease.

The apparent flame propagation rates, of both accelerating and decelerating flames within stratified methane-air mixtures caused by convective mixing of methane in air, were higher than those encountered for homogeneous quiescent conditions at the corresponding methane concentration in air.

Acknowledgements

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PUBLICATION NO. 2

"The Diffusivity of Methane into Air Within Vertical Tubes" (R. Petela and G. A. Karim); Proceedings of the 2nd ASME Fossil Fuel Combustion Symposium, January 14-18, 1989, Tokyo, Japan.

The Diffusivity of Methane into Air

Within Vertical Tubes

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ABSTRACT

An experimentally based effective diffusivity for methane is derived. Its application for predicting the concentration changes following the release of methane upwards into air at constant temperature and pressure within a vertical cylinder is then examined. Examples relating to the formation and dissipation of flammable mixtures following the release of methane into air are presented and discussed.

INTRODUCTION

Stratified atmospheres made up of combustible gases non-uniformly mixed with air are frequently encountered in various practical situations, including numerous industrial applications constituting a fire hazard. The formation of stratified mixtures, whether in various combustion chambers, around any fuel leaking off storage tanks or pipelines, or gas pockets in coal mine galleries are some examples. The development of a flame following ignition within such varying mixtures will depend on the stratification pattern formed mostly by diffusional processes. Molecular diffusion is often supplemented and combined with either natural or forced and laminar or turbulent convection. Much information is available in the open literature about the fundamentals of diffusion [e.g. 1-8], as well as the role of buoyancy effects [9,10]. However, there is still much need to provide further understanding of the formation of stratified mixtures by convective diffusion.

For example, the diffusion of the buoyant fuel methane in air, which is the main problem of interest to this contribution, involves the coefficient of interdiffusion as a function of temperature and pressure, and has been investigated both theoretically and experimentally [11,12]. Later on, other factors such as air speed, pressure differences [13], buoyancy effects [15,16,17], and ventilation rates [18] were examined. The effects of the density gradient in a system inherent in natural convention were also discussed [9,19,20]. Furthermore, buoyancy-driven flows were examined by Karim and co-workers [21,22] by developing one-dimensional models for convection diffusion and comparing the results to the corresponding experimental results.

It is evident throughout that modeling the formation of a stratified atmosphere, even for the simple cases and when approximate methods are employed, would involve inevitably complicated numerical methods [1,11]. In view of the complexity of the solutions and the associated phenomena involved, the application of an experimentally based effective diffusivity to replace the molecular diffusion coefficient is examined for the processes of the release of methane upwards into air at constant temperature and pressure [23,24,25]. This effective diffusivity concept was then used to interpret in a generalized manner the calculated concentrations of any gas diffusing into air or another gas in a one-dimensional system [26].

THE PROBLEM

The current paper presents some further quantitative observations on some practical aspects of the diffusional processes that can generate combustible mixtures, largely within tubes at constant pressure and temperature. For simplicity, the diffusional situations being considered are those assumed to take place along the tube shown schematically in Figure 1. The location of interest in the case of the one dimensional problem can be established by the axial coordinate, x, while for the two dimensional problem by 'x' and the radial coordinate 'r'. The tube is closed at the bottom ($x = 0$), but can be either closed or open at the top ($x = L$). A lower section of the tube having a length of h($0 \leq x \leq h$) and the remaining top part ($h \leq x \leq L$) are assumed to be filled with two different gases that are initially separated by a thin plate partition located at $x = h$. The process of diffusion is initiated between the two gases at $t = 0$, by removing the thin partition promptly (23). The local concentration 'c' of the diffusing gas at any location, x, becomes then a function of time, t, and location, x, (i.e. $c = c(x,t)$ or $c = c(x,r,t)$ for the two dimensional problem). If the densities of the two diffusing gases were different, then buoyancy effects may appear when the heavier gas overlays the lighter gas within the vertical tube. Of course, in the case of a horizontal tube with a sufficiently small diameter, d, the buoyancy effect becomes negligible.

The upward diffusion of methane into air was examined experimentally within a vertical circular transparent smooth tube of 63.5 mm diameter and 2.97 m length which was open at the top. Much of the experimental work was carried out with the lower section of the tube containing the methane (h) with a length of 203 mm. The mole fraction (c) of methane was established experimentally as a function of diffusion time, t, at three locations (x_1, x_2, x_3). This was achieved without disturbing the rapid diffusional process by using a specially developed ultrasonic gas analyzer [23]. Thus the transient concentration of the gas in air at any location was established effectively by measuring the apparent velocity of sound across a tube diameter at that location.

For the cases being considered here, the bottom part of the tube was initially filled entirely with methane (i.e. C_o = 100%), while the upper part of the tube was filled with pure air, at constant ambient temperature and pressure. Due to the strong buoyancy effects of methane in air, the measured values of the concentration of methane, of course was very significantly different from those that could have been obtained by calculations on the basis of entirely molecular diffusion with a constant coefficient of diffusion (D_M = 0.20 cm²/s), [3]. The differences between these two values for the concentration of methane are the subject of this contribution.

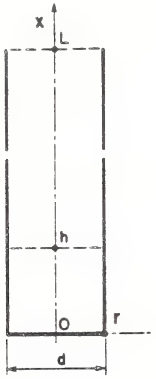

Fig. 1 - Schematic diagram of the circular cylinder being considered.

MOLECULAR DIFFUSION

The concentration of the gas following one-dimensional unsteady molecular diffusion within the tube of Figure 1, can be described as a volumetric fraction 'c' of the gas, assumed initially to be contained within the lower part of tube:

$$c(x,t) \tag{1}$$

'c' will be the solution of the following familiar differential equation for unsteady one-dimensional diffusion:

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \qquad 0 \leq x \leq L \tag{2}$$

where D is the coefficient of molecular diffusion, assumed usually to be constant.

The initial conditions associated with the case being considered are:

$$c(x,0) = c_o \qquad \text{for } 0 \leq x \leq h, \quad t = 0 \tag{3}$$

$$c(x,0) = 0 \qquad \text{for } h \leq x \leq L, \quad t = 0 \tag{4}$$

where c_o is the initial value of c, assumed here unless otherwise stated to be 1.0.

For both the open and closed top end cases, the following boundary condition applies:

$$\frac{\partial c}{\partial x} = 0 \qquad \text{for } x = 0,\ t \leq 0 \tag{5}$$

For the open ended tube at the top, the concentration 'c' at the exit of the tube is always zero [1,8],

$$c(L,t) = 0 \qquad \text{for } t > 0 \tag{6}$$

Hence, the differential equation (2) can be readily solved, for example, in terms of 'erf' functions.

It is also possible to solve the problem separately for the two regions 0 ≤ x ≤ h and h ≤ x ≤ L, by applying the Laplace transform method [24]. Due to different arguments of the 'erf' functions, the solution values of the concentration begin convergency from different initial values, but they converge ultimately to the same final solution value within the whole range (0 ≤ x ≤ L).

$$c = \frac{c_o}{2}\sum_{n=0}^{\infty}(-1)^n\left\{-\mathrm{erf}\left[\frac{2Ln-h+x}{2\sqrt{Dt}}\right] + \mathrm{erf}\left[\frac{2Ln+h+x}{2\sqrt{Dt}}\right] + \mathrm{erf}\left[\frac{2L(n+1)-h-x}{2\sqrt{Dt}}\right] - \mathrm{erf}\left[\frac{2L(n+1)+h-x}{2\sqrt{Dt}}\right]\right\} \tag{7}$$

in which n > 4 is usually sufficient to produce an acceptable accuracy.

Applying the method of separation of variables for the whole range, 0 ≤ x ≤ L, the following solution can be obtained:

$$c = \sum_{n=1}^{\infty}\frac{2\,c_o}{(n-0.5)\pi}\sin\left[\frac{(n-0.5)\pi h}{L}\right]\cos\left[\frac{(n-0.5)\pi x}{L}\right] \exp\left[-D\frac{(n-0.5)^2\pi^2}{L^2}\,t\right] \tag{8}$$

The solution values from equations (7) and (8), obviously would be the same. However, for the case of the tube with the closed top, the following condition applies:

$$\frac{\partial c}{\partial x} = 0 \qquad \text{for } x = L, \qquad t \leq 0 \tag{9}$$

Applying the separation of variables method for the whole range, $0 < x < L$, the following solution can be obtained:

$$c = \frac{c_o h}{L} + \frac{2c_o}{\pi} \sum_{n=1}^{\infty} \frac{1}{n} \sin\left(\frac{n\pi h}{L}\right) \cos\left(\frac{n\pi x}{L}\right) \exp\left(-D\frac{n^2\pi^2}{L^2} t\right) \tag{10}$$

The exponential expression in time of equation (10) controls strongly the final result as the other sine and cosine expressions can be maximum and equal to unity. Assuming the 'ε' is the value of the last 'nth' term taken into account at an assumed degree of accuracy, then the following condition can be stated:

$$\exp\left(-\frac{n^2\pi^2 Dt}{L^2}\right) > \varepsilon \tag{11}$$

and the number of the series terms taken into account is:

$$n > \text{int}\left(\sqrt{-\frac{L^2 \ln\varepsilon}{\pi^2 D\, t}}\right) + 1 \tag{12}$$

where 'int' converts the value of the argument to type integer by truncation. The problem is illustrated by the diagram, Figure 2, for an assumed value of $\varepsilon = 10^{-5}$, and with 'n' as a real number. It can be seen that larger numbers of terms are needed for shorter diffusion time and longer tubes.

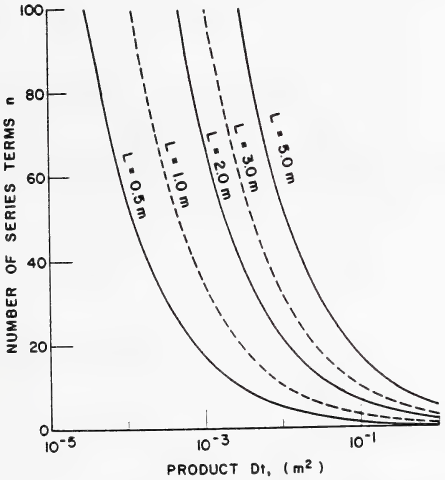

Fig. 2 - Variation of number of terms needed with the product Dt to obtain a certain degree of accuracy for a range of tube lengths.

Equations (10) and (7) or (8) are used to calculate the concentrations in the closed and open top tube end, respectively, under the same conditions. Figure 3 shows that in the initial period of the diffusional process for both cases with closed (solid line) and open (broken line) ends, the concentration curves are the same up to the time when the open end of tube begins to influence markedly the process of diffusion within the tube. This characteristic time, shown in Figure 3 with the dashed line, expectedly, is greater the further distance, x, from the open end considered. Moreover, it can be seen that for the open ended tube, the nearer the relative location to the fuel section, the higher the fuel concentration and the earlier it subsides.

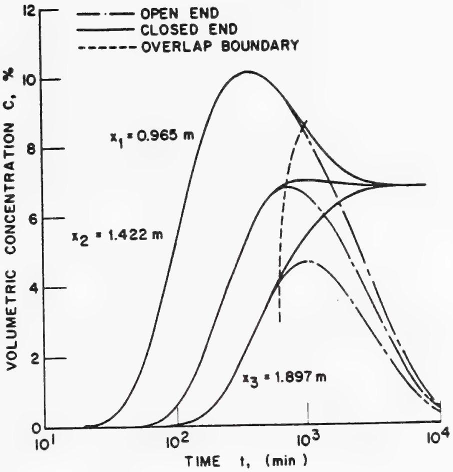

Fig. 3 - Variation of the concentration of methane with time at three locations along the tube for both open and closed upper ends. The time boundary when the open end begins to influence the diffusion is also shown.

Using equations (8) and (10), the values of the dimensionless characteristic time, D_{t_o}/L^2, after which the gas concentration for the open and closed ends cases differ by more than 1%, is shown in Figure 4 as a function of the dimensionless length ratios x/h and h/L. It can be seen that longer times are required for relatively smaller size fuel sections. Prior to this time, the concentration curves for both open and closed tube end, are essentially the same.

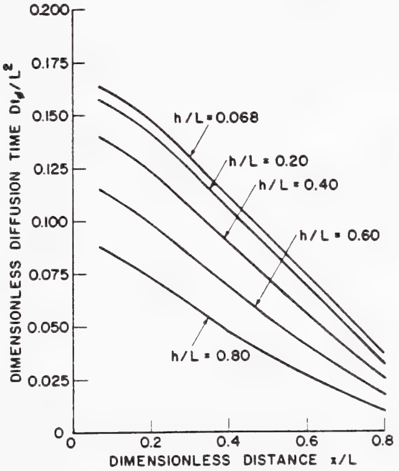

Fig. 4 - Variations of the dimensionless diffusion time with dimensionless distance when the gas concentration for the open and closed ends cases differ by more than 1% for a range of h/L values.

EFFECTIVE DIFFUSIVITY

Due to the strong buoyancy effects of methane diffusing upwards into air within the open tube of Figure 1, the measured methane concentrations as shown in Figure 5, are shifted significantly earlier in time, almost by two orders of magnitude, in comparison with the corresponding calculated values for molecular diffusion. However, the shape of the curves tends to be similar. Similarly, the experimental curves overlap initially for both the open and closed end cases up to the line shown.

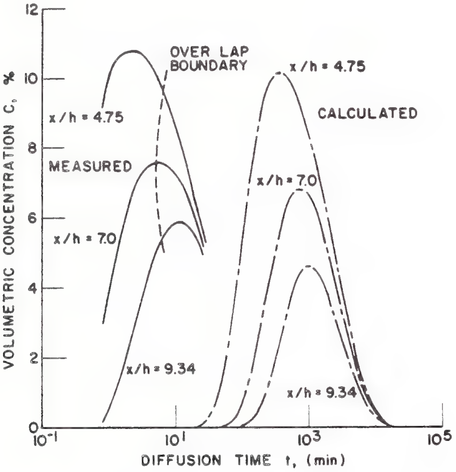

Fig. 5 - Variation of experimental and calculated concentrations of methane with time at three locations. The calculations are carried out according to molecular diffusion.

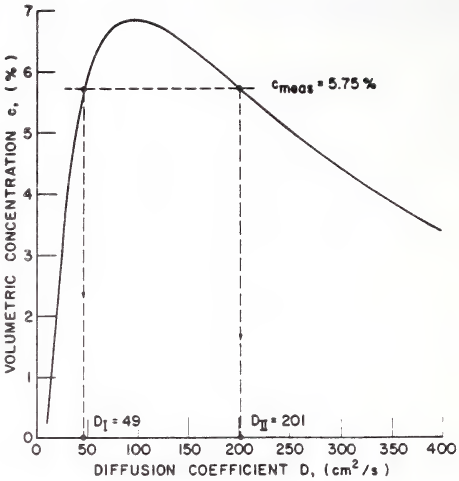

Fig. 6 - Variation of the calculated volumetric concentration with diffusion coefficient for a certain location and time.

The actual diffusion of methane in air seen in the tube under the influence of buoyancy, is complex, since it can involve unstable and turbulent processes. Accordingly, for convenience and when some experimental data are available, it can be conceived as being equivalent to a molecular diffusional process with an appropriately different diffusional coefficient 'D' that has been described as an "effective diffusivity", [24]. Obviously, this diffusivity will be varying widely with respect to time and operating conditions. A local instant value could be determined in principle for any concentration molecular diffusion solution, e.g. equation (7), (8) or (10). For a typical example for the time t = 96 s, three measured concentrations, $c_1 = 10.75\%$, $c_2 = 5.75\%$ and $c_3 = 1.7\%$, are obtained for the locations $x_1 = 0.965$ m, $x_2 = 1.422$ m, and $x_3 = 1.879$ m, respectively. Equation (7) is used to generate the corresponding dependence of c = c(D) on the value of the effective diffusion coefficient, as shown in Figure 6 for location x_2. It can be seen that the value of the experimentally measured concentration ($c_2 = 5.5\%$), can be achieved with two widely different values of effective diffusivity, $D_I = 49$ cm²/s and $D_{II} = 201$ cm²/s. The additional experimental points obtained at other locations at the same time, as shown in Figure 7, indicate that the lower value of effective diffusivity, $D_I(x_2,t_j)$, needs to be taken for calculating the actual fuel concentration at t = 96 s. Similarly, using equation (7) for the corresponding experimental data at location x_3, one can fit, for example, the value $D_I(x_3,t_j) = 37.5$ cm²/s.

This approach of finding values of the local instant effective diffusivity was applied to interpret the measured concentrations by plotting the dimensionless factor D_I/D_M, of the effective diffusivity D_I and the molecular diffusivity, $D_M = 0.2$ cm²/s, as a function of time for constant values of the dimensionless location x/L.

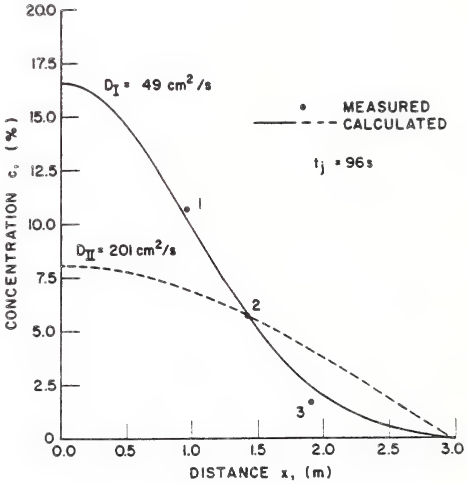

Fig. 7 - Calculated values of methane concentration versus distance for two values of the diffusion coefficient. Some experimental points obtained are also shown.

Considering the physical nature of the buoyant convection diffusion phenomena, the effective diffusivity can be assumed to be a function of primarily two variables. Since the buoyancy of the methane in the tube in the absence of temperature differences is driven by the density difference of the diffusing gases, then one of these variables could be the dimensionless density:

$$\left(\frac{\rho_A - \rho}{\rho_A - \rho_M}\right)_T$$

in which ρ_A and ρ_M denote the density of air and methane, respectively, and ρ denotes the changing density of the local mixture of air and methane. This expression can be shown for any constant temperature to be equal to the local mole fraction 'c' of methane in a mixture. Thus the percentage concentration 'c' can be used as one of the controlling variables.

The transport of any species is actually driven by the difference in concentration of this species in space so that the gradient $\frac{\partial c}{\partial x}$ can be used as the second variable. Thus, optimization of the exponents for the corresponding experimental data leads to the following formula for the effective diffusivity of methane upwards into air at constant ambient temperatures and pressure:

$$D = D_M \; [1 + (\frac{\partial c}{\partial x})^{2.3672} + c^{0.00024}] \tag{13}$$

It can be seen that the above expression indicates that it is virtually independent of concentration since:

$$c^{0.00024} \approx 1.$$

Moreover, because:

$$(\frac{\partial c}{\partial x})^{2.3672} >> 2$$

the following formula could be proposed for the effective diffusivity to be used in approximate calculations of the transient concentrations of methane according to equation (2):

$$\frac{D}{D_M} \propto (\frac{\partial c}{\partial x})^{2.37} \tag{14}$$

in which

$$1 \leq (\frac{\partial c}{\partial x}) \leq 10 \text{ \%/m} \quad \text{and} \quad 60 \leq t \leq 390\text{s}.$$

The standard deviation between calculated values using equation (14), and the corresponding measured values of the concentration is $S = 0.20345$ which indicates that $c \pm 0.20345\%$. Expression (14) can be rearranged in terms of the dimensionless distance $\eta = x/L$ to:

$$\frac{D}{D_M} \approx 4120 \; (\frac{\partial c}{\partial \eta})^{2.37} \tag{15}$$

in which

$$0.03 \leq \frac{\partial c}{\partial \eta} \leq 0.3 \quad \text{and} \quad 60 \leq t \leq 390 \text{ s}.$$

and where the magnitude of 'c' denotes the mole fraction of methane in the mixture.

For the same set of experimental data used above, the dimensionless product of the concentration gradient and the distance coordinate was tried in another approximate formula:

$$D = D_M \; [1 + 78.5(x \, \frac{\partial c}{\partial x})^{0.002}] \tag{16}$$

in which

$$7 \leq \frac{\partial c}{\partial x} \leq 14.5 \text{ \%/m}.$$

If equation (16) is used, with a standard deviation between calculated and measured values of concentration $S = 0.315$, for $0.95 \leq x \leq 1.35$ m and for $48 \leq t \leq 96$ s, then the effective diffusivity value becomes:

$$D/D_M = 3.025 - 7.25t + 5.78xt - 36.67x \tag{17}$$

CALCULATIONS FOR THE GENERATION OF FLAMMABLE ZONES

An important practical problem is the assessment of the fire and explosion hazards arising from the release of a combustible gas into air [25]. For these considerations, a knowledge of the concentration changes of the components of the mixture with time and space is needed. The analytical procedure described earlier relating to the convection diffusion of methane into air can be employed to establish more realistically for any configuration whether flammable concentrations are reached. It would be possible for any location to establish the time requirements for the formation of combustible mixtures and for their subsequent subsistence to levels that would render them non-flammable, either temporarily or permanently.

A widely used criterion in such an assessment is the flammability limit, whether lean or rich. For a mixture of methane and air at normal ambient conditions, these limits can be assumed to be 5.5% and 14.0% of methane by volume for the lean and rich limits, respectively. Using experimentally obtained data for the open tube system of Figure 1, with some extrapolation, it was possible to plot the curves shown in Figure 8 of some constant methane concentrations close to the lean limit ($C = 4.0\%$, 5.0% and 6.0%) both for the initially increasing concentration and then after the lapse of much time for the decreasing concentrations as a function of location and time. It can be seen that for the case considered, a flammable mixture of methane and air can be formed in a matter of less than a few minutes at a location of $x = 1.5$ m. However, it would take nearly in excess of a half hour for the mixture at this location to become non-flammable.

It was found that considerable regions of such experimentally derived curves can be generated with relatively good accuracy using equation (7) while employing appropriately chosen constant values of the effective diffusivity. The use of variable values based on formulations such as those of equation (15) require considerable amount of mathematical manipulations. As shown typically in Figure 8, a $D = 50$ cm^2/s is used for the earlier increasing concentrations curves and a $D = 18$ cm^{2}S for the later decreasing curves. For comparison the decreasing curve of 5.0% methane generated while using a $D = 50$ cm^2/s is also shown. Figure 8 also shows the region in which the assumption of a one-dimensional decreasing curves. For comparison the decreasing curve of 5.0% methane generated while using a $D = 50$ cm^2/s is also shown. Figure 8 also shows the region in which the assumption of a one-dimensional diffusion becomes less certain (27). This is largely associated with extremely early times and in the neighbourhood of the initial fuel-air interphase.

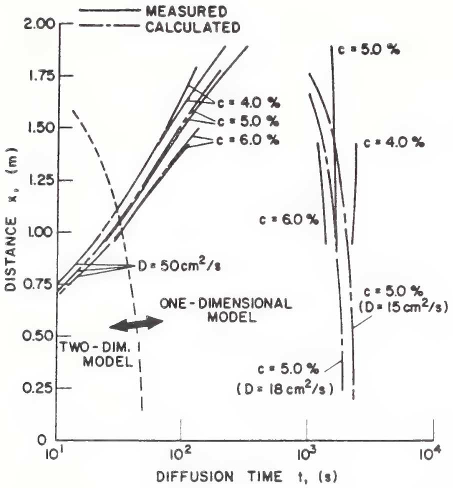

Fig. 8 - Various concentration profiles of methane in air around the lean limit.

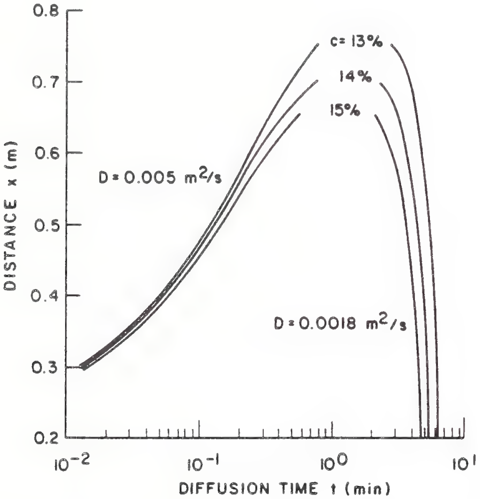

Fig. 9 - Typical calculated concentrations for three rich mixtures in the tube of Fig. 1 as a function of distance and time when using two different effective diffusivity values for the upward diffusion of methane into air.

Similarly, using effective diffusivity values of $D = 50\ cm^2/s$ for diffusion times of shorter than 60 s and $D = 20\ cm^2/s$ for times greater than 60 s, three constant methane concentration curves (c = 13.0, 14.0 and 15.0%) close to the rich limit are shown typically in Figure 9. Using data for the curve of c = 14% to represent the rich limit and for the curve of c = 5.5% to represent the lean limit, Figure 10 shows typically the characteristic regions of flammability as a

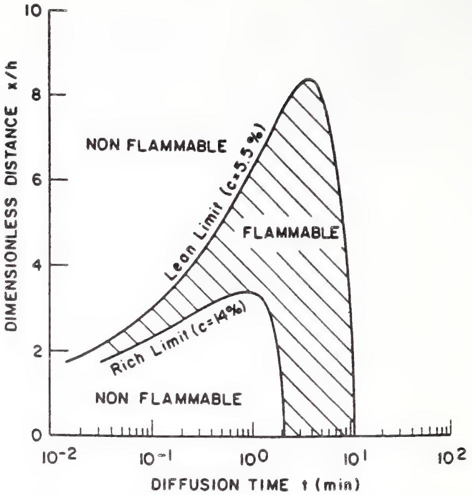

Fig. 10 - Typical calculated concentrations for the flammable region during upward diffusion of methane into air in the tube of Fig. 1 as a function of distance and time when using an effective diffusivity value.

function of diffusion time and dimensionless distance. Similar regions obtained entirely on the basis of the molecular diffusion of methane in air [26] would have produced totally erroneous estimate of the time required to produce a flammable mixture at any specific location.

ACKNOWLEDGEMENT

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NOTATION

c	molar concentration
D	diffusion, coefficient (D_M) and effective diffusivity (D_I)
h	length of fuel section
L	tube length
n	number of terms
r	radius
S	standard deviation
t	time
x	linear distance
η	dimensionless distance (x/L)
ρ	density
ε	value of nth term

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PUBLICATION NO. 3

"Examination of the Combustion of a Fuel Jet in a Homogeneously Premixed Lean Fuel-Air Stream" (G. A. Karim, M. Kibrya, R. Lapucha and I. Wierzba); Proceedings of the International Fuels and Lubricants Meeting and Exposition of the SAE, Portland, Oregon, October 10-13, 1988, Paper No. 881662.

881662

Examination of the Combustion of a Fuel Jet in a Homogeneously Premixed Lean Fuel-Air Stream

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ABSTRACT

There are numerous situations in a wide range of engineering applications involving combustion devices where the combustion of a fuel jet takes place in flowing streams containing varying proportions of a fuel homogeneously premixed with the surrounding air. Such applications can be found, for example, in dual fuel engines and in some gas turbine combustors.

The paper describes some of the findings of an experimental investigation, supported by some analytical modeling, of the combustion of a circular gaseous fuel jet within lean homogeneous mixtures of various gaseous fuels and air. The nature of the combustion process of the pilot fuel jet, flame spread characteristics and limits within the surrounding moving atmosphere were considered in terms of the fuels used for the jet and the surrounding atmosphere and in terms of the jet discharge and surrounding stream flow characteristics.

THERE ARE NUMEROUS SITUATIONS involving industrial combustion devices where the combustion of a fuel jet takes place in a non-quiescent environment that itself is potentially combustible. Examples can be found in engines whether of the dual fuel compression ignition type or precombustion chamber engines where a flaming jet ignites the bulk of the homogeneously mixed fuel-air charge. Other examples can be cited in relation to gas turbine combustors, especially those of the prevaporizing combustors, or in furnaces employing dual fuel systems. The mixing and combustion processes involved in such devices are much too complex and some of their details remain yet uncertain. They involve the simultaneous combustion of two fuels in varying proportions in diffusion and premixed modes within a highly turbulent environment. There is a need for a better understanding and control of such processes, especially if the enormous resources of alternative fuels such as those of natural gas are to be utilized efficiently, conveniently and economically in conventional combustion engines and devices. Moreover, it has been reported relatively recently that the stability of a jet diffusion flame can be improved significantly when its surroundings contain some fuel mixed with the air (1)*. Associated with this approach is the potential for modifying the temperature and concentration fields of any jet diffusion flame through the presence of small concentrations of fuel in its surroundings (2,3).

Accordingly, the paper reviews the results of a laboratory investigation into the combustion of a relatively small gaseous fuel steady jet in a steady low velocity unidirectional stream of very lean homogeneous fuel-air mixtures. These conditions though appear to be quite far removed from those normally encountered in the combustion chamber of gas turbines or dual fuel engines, they can nevertheless in view of the complexity of the processes involved provide useful pointers that will aid in our understanding of the complex processes found in industrial combustors.

SOME THEORETICAL CONSIDERATIONS

The concentration of a fuel at any point within an axisymmetric cylindrical steady fuel jet issuing into an air environment depends on the rate of entrainment of the air by the issuing jet. The resulting axial and radial fuel concentration profiles can be calculated by employing well established theoretical procedures (e.g.4,5). Hence, for any fuel and set of operating conditions, the locations of the lower flammability, stoichiometric or the higher flammability limit enveloping zones can be established. Furthermore, by considering the flame to be associated with the combustion of a certain concentration of fuel in air, the possible location of the flame envelope can be obtained.

When a small amount of auxiliary fuel is mixed with the surrounding air in the vicinity of the issuing fuel jet, the jet will entrain the

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*Numbers in parentheses designate references at end of paper.

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surrounding fuel along with the air. Therefore, although the general shape of the fuel concentration distribution will remain essentially the same, the concentration of the total fuel at any point will be increased due to the induction of the surrounding fuel with the air while the concentration of the air reduced by the same amount, as shown schematically in Fig. (1). Consequently, the fuel-air ratio at that point will become correspondingly higher and the stoichiometric region would move outwardly towards a larger radius. This results in an increase in the overall size of the flame at every section downstream of the nozzle, both in width as well as length. This increase will be particularly prominent at regions away from the discharge point of the jet. Similarly is the case for the flammability limit zone surrounding the jet flame. Since the rate of fuel consumption by a flame is given by the product of its surface area and the local burning velocity, it becomes possible to produce more thermal energy from the same jet by burning relatively more fuel. This is particularly the case if the local burning velocity of the fuel mixture at the flame surface is increased due to the presence of the two fuel components. However, since their relative concentrations would vary along the flame surface, the local burning velocity would also change along the flame surface.

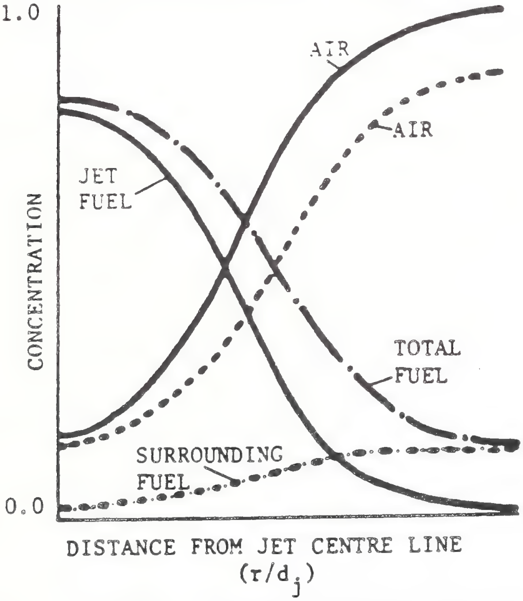

Fig. 1 Schematic representation of the concentration profiles at a given cross-section within a fuel jet issuing in an atmosphere of air or fuel-air homogeneous mixture.

It is the common practice for convenience of analysis to assume that the diffusion flame will be located along the stoichiometric mixture envelope. However, over the wide range of fuel discharge rates normally encountered, a diffusion flame may exist in practice somewhere within the region bounded by the lean and the rich flammability limits mixtures envelopes. It has been shown experimentally by Karim and Kibrya (6) that when high velocity jet flames near the blowout limits are involved, the resulting flame would be located much nearer to the lower flammability limit envelope than to the corresponding stoichiometric envelope.

Computations were made, on the basis that the flame is associated with the lower flammability limit envelope, as well as for comparative purposes when based on the traditional assumption that the flame is located along the stoichiometric mixture envelope. Fig. (2) shows typically the computed lower flammability mixture zone for a cold turbulent gaseous fuel jet issuing into an environment containing various concentrations of an auxiliary fuel (e.g. methane, ethane or propane), as well as the corresponding computed stoichiometric zone, Fig. (3). In these relatively simple calculations, the jet is assumed to be cylindrical, steady asymmetrical and at the same temperature as the environment. From such plots it can be seen that both the width and the length of the resulting flame will increase very markedly as the concentration of the auxiliary fuel in the jet surroundings is increased. These figures tend to confirm also that a significant increase in the total burning rate of a jet burner would be accomplished when adding small concentrations of an auxiliary fuel to the surrounding air. The increase in the burning rate would be small at the beginning when very small concentrations of the auxiliary fuels are employed. The increase becomes very substantial when the concentration of the auxiliary fuel in the surroundings approaches the value that allows flame pro-

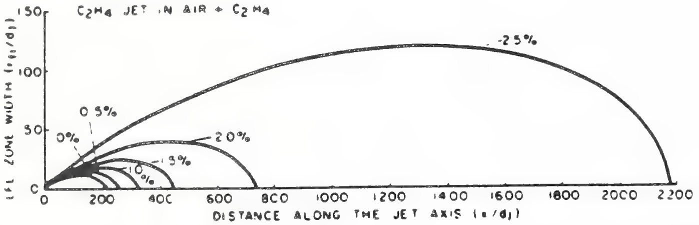

Fig. 2 Boundaries of the lower flammability limit zone (LFL) of a turbulent methane jet issuing into a co-flowing low velocity stream (0.22 m/s) of air and methane for various volumetric concentrations of the surrounding fuel (%) methane in the air.

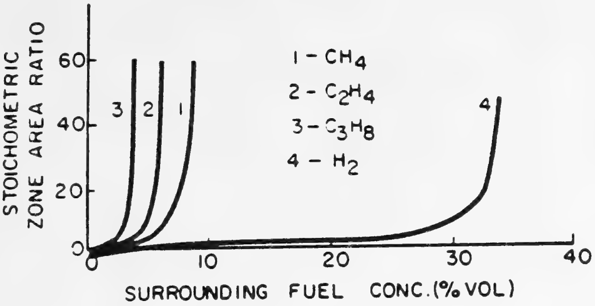

Fig. 3 The ratio of the flame surface areas based on the stoichiometric zone when an auxiliary fuel is present in the surroundings relative to a surrounding of air only as a function of the volumetric concentration of the auxiliary fuel for four auxiliary fuels with methane as the fuel jet.

pagation from the fuel jet flame to the whole surrounding mixture. Accordingly, to ensure that the auxiliary fuel in the surrounding air will be consumed by a propagating flame from the jet discharge, the auxiliary fuel concentrations need to be kept throughout well above this limiting concentration so as to avoid having the combustion process confined to the regions of the fuel jet flame only. This is relevant for example to the common problem encountered with dual fuel operation at very light load where poor fuel utilization results from incomplete flame propagation within the fuel-air mixture from the pilot combustion zone.

It would be expected that for a small size jet, the combustion of the surrounding fuel at very lean mixtures would be extremely small and confined to the immediate neighborhood of the main jet. As the size of the jet is increased, the length of the flame increases (7) and the combustion of the auxiliary fuel increases proportionately to the area of the resulting main flame. However, when the surrounding stream contains fuel concentrations beyond a critical concentration that corresponds to the effective flammability limit, then obviously even a small size jet will be quite adequate to initiate combustion over the whole mixture region through a propagating flame originating from the jet pilot flame.

EXPERIMENTAL SETUP

The experimental setup employed in this study of the combustion of a model jet flame in parallel flowing streams that may contain some additional mixed fuel was described previously [(7), (8)]. The apparatus, Fig. (4), was a vertical steel combustor of 150mm diameter and 400mm height. It was open at the top and fitted with a special glass window for viewing the flame. A sharp-edged nozzle located vertically along the centre line of the combustor was used to produce a small diffusion pilot flame. The surrounding premixed fuel-air stream flows from the bottom of the combustor. Before entering the combustor, the flow passes through a settling chamber, a perforated cone, a set of stainless steel wire mesh, and a honeycomb flow straightener to produce a uniform velocity distribution at the inlet section of the combustion chamber. The wire mesh also acts as a flame arrestor in case of flame flashback through the honeycomb.

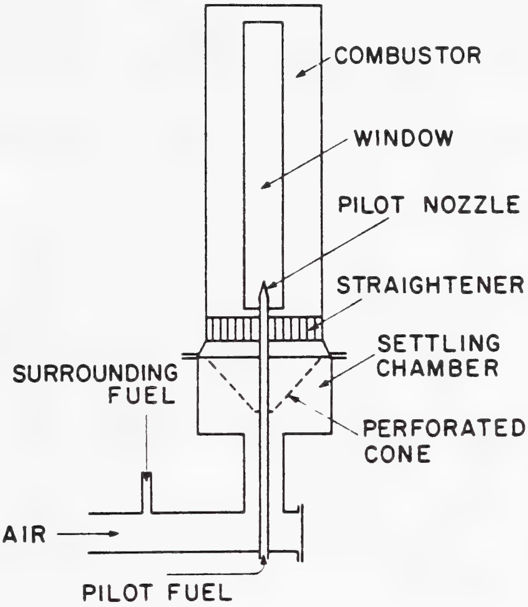

Fig. 4 Schematic diagram of apparatus.

The various fuels tested were supplied from high pressure cylinders, while air was taken from the compressed air supply of the laboratory. Much of the testing was carried out with commercial methane (typically composed of 94.7% CH_4, 3.7% C_2H_6, 0.7% C_3H_8, 0.2% C_4H_{10}, 0.4% C_5H_{12}, and 0.3% N_2). The fuels in the surrounding stream were in turn methane, propane, ethylene, or hydrogen. In these tests the surrounding stream velocity was kept relatively low and ranged from 0.025 to 0.60 m/s. The volume flow rates of the fuels and air were measured using choked nozzles. A hot-wire anemometer was used to establish the velocity profile of the jet and the surrounding streams. The analysis of the combustion gases was carried out largely by means of Beckman NDIR analyzers with a water-cooled stainless steel 2.5mm diameter probe used for sampling. The behaviour of the flame system was video-recorded throughout for subsequent analysis. For example, the angle of the propagating flame front at the moment of flame flashback from the jet flame into the surroundings was measured using such video records.

The limits for flame spread into the surroundings from the pilot jet flame were determined either by increasing the fuel concentration in the surroundings while keeping the stream velocity constant or by keeping the flow rate of the fuel constant, and varying the stream velocity. This later approach provided also varying velocities of the surrounding stream.

THE BLOWOUT LIMIT OF A JET DIFFUSION FLAME

When the jet fuel discharge rate is gradually increased, typically the diffusion flame comes to a point when it lifts from the tip of the nozzle while remaining essentially stable as a detached turbulent flame resulting in vigorous noisy combustion. With a further increase in the fuel discharge rate, the lift-off height increases until it reaches a limiting stage when flame blowout is encountered somewhat abruptly.

The introduction of some fuel in the surroundings represents a situation whereby a premixed ultra-lean fuel-air stream is flowing over and past a steady jet diffusion flame. Combustion can be extended somewhat into the premixed stream along the surface of the pilot flame, producing a thin envelope of lean-premixed combustion zone immediately around the main flame. The envelope thus produced provides additional thermal energy to the diffusion flame. The air normally entrained by the jet diffusion flame gets preheated and carries additional active radicals into the flame as it passes through this envelope. Also, since the flame lifts up before blowout is reached, some premixing of the jet fuel and the surrounding fuel takes place at the base of the flame. This would increase the local equivalence ratio, resulting in an increase in the local flame speed, thus enabling the flame to withstand a higher discharge velocity.

The surface area and the effective thickness of the diffusion flame also increase with the introduction of fuel in the surrounding. This increase in the flame surface area and the additional thermal energy input by the surrounding flame envelope enable the diffusion flame to be maintained with a much higher discharge velocity. Fig. (5) shows typically the volumetric blowout limits of a methane circular jet diffusion flame discharging in a co-flowing surrounding stream of air containing, in turn, some hydrogen, propane, ethylene or methane homogeneously mixed with it. It is evident that a significant extension of the blowout limit can be produced through the presence of these small concentrations of the surrounding fuel in the stream (9). The concentration of the auxiliary fuel in the surroundings is represented by the equivalence ratio of[1] the homogeneously premixed surrounding stream.

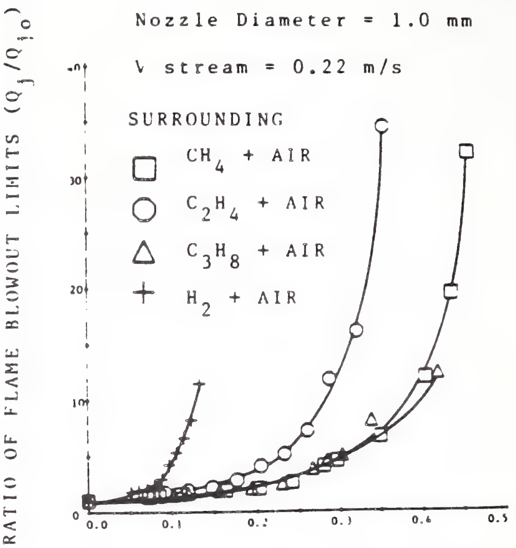

Fig. 5 The ratio of the flame surface areas (A_{fs}/A_{fa}) based on the flammability limit zone when an auxiliary fuel is present in the surrounding relative to a surrounding of air only as a function of the auxiliary fuel volumetric concentration for four auxiliary fuels with methane as the fuel jet.

This apparent extension to the blowout limit of the jet diffusion flame, as the concentration of the fuel in the jet surroundings is increased, appears to be proportional to the estimated increase in the surface area of the enveloping flame (6), whether when based on the stoichiometric mixture zone or the lean limit mixture zone, Fig. (6).

In a setup such as the one being described here, since the surrounding air stream contains some premixed fuel, flame propagation through the stream from the diffusion jet flame becomes an important consideration from operational and safety points. Hence, the maximum amount of fuel concentration that can be premixed in the surrounding air stream, and consequently the potential for extending the jet diffusion blowout limit, will be limited by the onset of flame propagation through the whole surrounding stream.

The extension of the blowout limits of the jet diffusion flame for a given fuel in the surrounding was then related to the flashback concentration limit of the fuel in the stream. It was observed that for any set of operating conditions, when the concentrations of fuel in the surroundings are expressed relative to the corresponding flashback concentration of the fuel, the observed data can be reduced to a single curve, irrespective of the type of surrounding fuel employed, as shown in Fig. (7). This indicates that the degree of extension of the blowout limit is determined by how close to flashback the surrounding flame is, rather than the absolute concentration of the auxiliary fuel itself (9).

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[1] The equivalence ratio is the fuel to air ratio relative to the corresponding stoichiometric ratio.

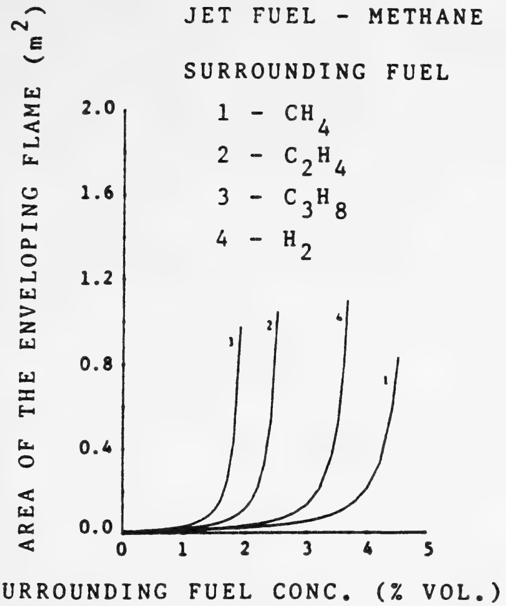

Fig. 6 Computed variation of surface area of the enveloping flame with an increase in concentration of fuel in the surrounding stream (9).

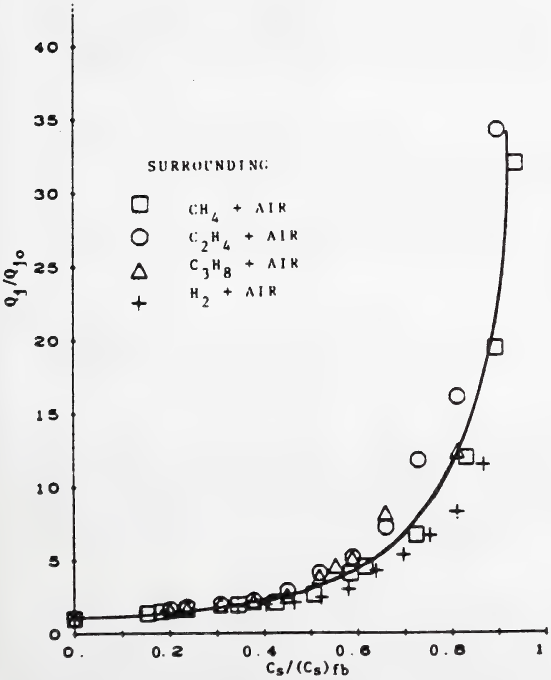

Fig. 7 Ratio of blowout limits of a methane jet diffusion flame as a function of the surrounding fuel concentration expressed as a fraction of flashback limit.

IGNITION AND FLAME SPREAD IN THE PRESENCE OF THE PILOT FLAME

In order to examine the mode of ignition and flame spread into a very lean homogeneously mixed fuel-air stream from a steady pilot jet flame, the fuel flow rate to the pilot was kept deliberately small and constant. As some fuel was introduced into the surrounding stream, combustion was initiated along the periphery of the pilot flame, even at very small concentration. Fig. (8) shows the variations of the pilot flame with increasing the fuel concentration in the surrounding stream. In the case of the fuel volumetric fraction, FVF, in the surrounding was zero, Fig. (8a), the pilot flame can be seen to be a typical jet diffusion flame for a very small discharge Reynolds number. The pilot zone is surrounded by a strong, easily visible, light-blue combustion zone and a very weak light blue afterburning region. As some fuel is introduced into the surrounding stream, a new zone appeared around the pilot flame, Fig. (8b). The pilot zone scarcely varies its appearance under these new conditions since enough oxygen probably exists after the surrounding mixture flame zone. With a further increase in the fuel concentration of the surrounding stream, the afterburning region is increased, and the top of the combustion zone, Fig. (8b-c), becomes a visible rich yellow-orange. This rich combustion zone grows as the concentration of the fuel is increased. Fig. (8d) shows schematically the pilot flame zone just before the flame flashback. These rich and strong combustion zones and the afterburner region disappear after flame flashback, Fig. (8e).

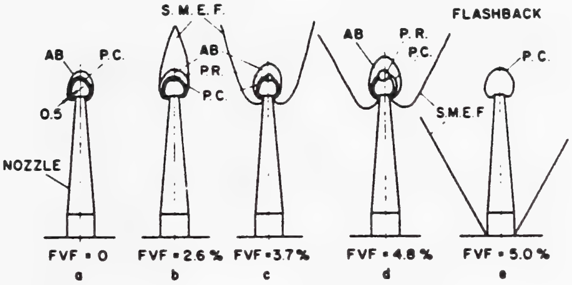

Fig. 8 Variation of the pilot flame zone; Vs = 0.23 m/s, do = 1.5 mm, CH_4 pilot fuel, Rej 20; AB, after burner zone, P.C., mixture flame front; P.R., pilot combustion rich zone.

Fig. (9) shows the schematic representation of the flame propagation from the pilot flame into the surrounding stream. With an increase in the concentration of the fuel, the surrounding mixture flame zone becomes wider and higher. The tip of the flame opens up for a certain concentration of the fuel, Fig. (9j), and V-shaped flames are produced. Increasing the fuel concentration further will eventually produce a critical "flashback angle," Fig. (9, 1). Increasing the concentration

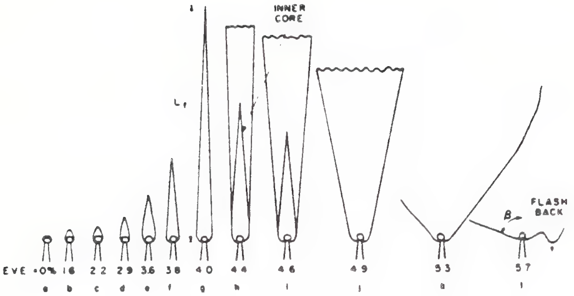

Fig. 9 Schematic representation of the flame propagation from the pilot flame into the surrounding stream; Vs = 0.06 m/s, CH_4 pilot fuel, CH_4 surrounding fuel, do = 1.5 mm.

beyond this point causes the flame to flashback onto the base of the combustor and propagates against the stream. It was observed that this critical angle between the leading edge of the flame front and the vertical axis of the combustor at which the flame flashes back, was normally different for different fuels. The angle was smallest for hydrogen and largest for propane. For the same fuel, an increase in the surrounding velocity caused a decrease in the critical angle.

Fig. (10) shows the critical angle of the flame front for methane, ethylene, and propane. Some attempts to measure the critical angle for hydrogen failed because of the dimness of the hydrogen flame for the corresponding concentration. A burning velocity can be calculated for an idealized model of the flame shape at flashback on the basis of the flame angle measured and the local stream velocity. Fig. (11) shows the

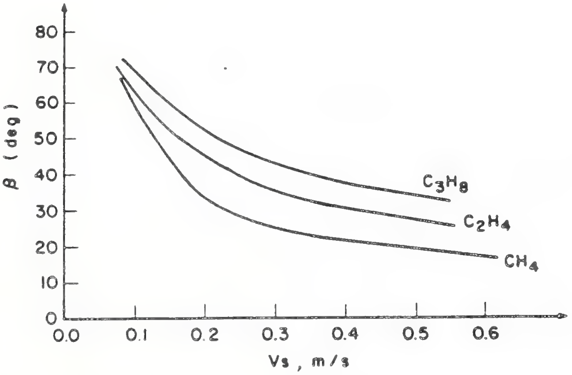

Fig. 10 Comparison of the variation of the front flame angle at the beginning of flame flashback for various hydrocarbon fuels vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5 mm.

variation in this calculated burning velocity. It can be seen that the burning velocity for propane is greater than that of ethylene, which is, in turn, greater than that of methane. For low surrounding velocities, Vs < 0.1, the differences between the burning velocities are comparatively small, while for Vs < 0.06, there is virtually only one burning velocity for those three hydrocarbons. This is consistent with observations of the burning velocities in limiting homogeneous mixtures (10,11).

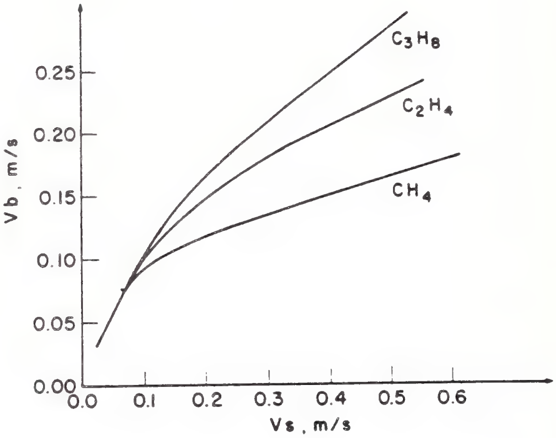

Fig. 11 Comparison of the variation of the burning velocity at the beginning of flame flashback for various hydrocarbon fuels vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5 mm.

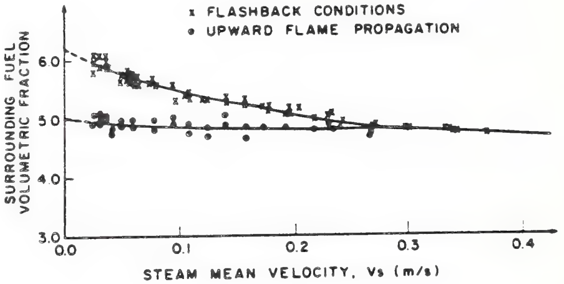

Fig. 12 Comparison of the variation of the fuel volume fraction in the stream at flame flashback and at flame upward propagation for CH_4 vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5 mm.

It was observed that for some fuel concentrations before flame flashback started, large portions of the flame tore off from the main surrounding flame, to be carried upwards and spread through the stream. This is analogous to conditions associated with the lean upward flame propagation limit. Fig. (12) shows the lean limit of this upward propagating flame in comparison to the lean flame flashback limits, both for CH_4 fuel. It is evident that at leaner mixture concentration, partial flame propagation along the stream takes place leading to partial oxidation of the fuel. However, as the concentration is increased further to another critical concentration, the flame propagates throughout and against the stream. Of course, as the stream velocity is increased, the difference in these two limiting concentrations decreases until a stream velocity is reached, when it disappears as the contribution of natural convection to flame spread becomes less significant.

The flashback of the flame did not occur at the same time over the entire region of the combustor. It should be noted that as the fuel concentration in the stream was increased gradually, the flame first started flashing back along the pilot flame nozzle for the surrounding stream velocities, Vs, of less than 0.15 ($Re \approx 1500$). The angle of the flame front was constant during this flashing back, Fig. (13a). For Vs < 0.1 m/s, the flashback of the flame started first far from the pilot flame nozzle, Fig. (13b,c) while for 0.15 < Vs < 0.1 m/s it started in a way either similar to the case of Fig. (13a) or of Fig. (13b,c). The flashback of the flame for surrounding stream velocities Vs < 0.05 m/s, and especially for heavier hydrocarbons (C_3H_8), proceeded in a relatively longer time with the flashback occurring after the flame began oscillating.

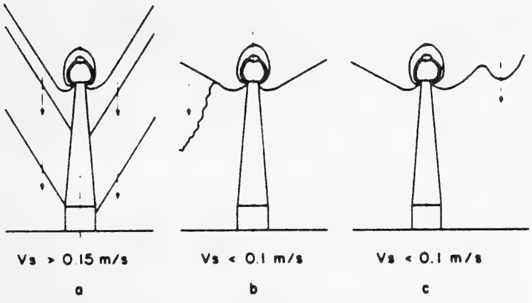

Fig. 13 Schematic representation of the flame flashback
a) flame flashback for Vs > 0.15 m/s;
b,c) flame flashback for Vs < 0.10 m/s.

Fig. (14) shows typically the concentration profiles of CH_4, O_2, CO_2, and unburned CH_4 at a specified distance above the nozzle for a combustible mixture with 5.1% methane. The efficiency of the combustion of the very lean mixture tends to be high and probably above 90%.

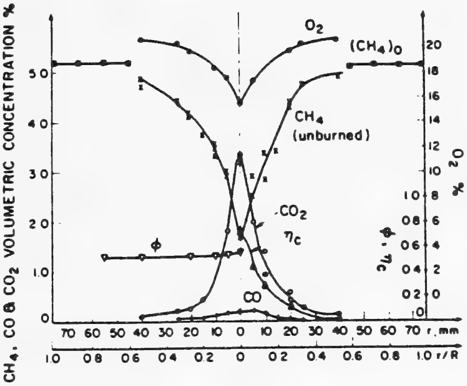

Fig. 14 Volume fractions of unburned CH_4, CO, CO_2, and O_2 vs. radial distance; initial volume fraction of CH_4 in the surrounding stream (CH_4) = 5.1%, Vs = 0.06 m/s, CH_4 pilot flame, do = 1.5 mm, h = 22 mm.

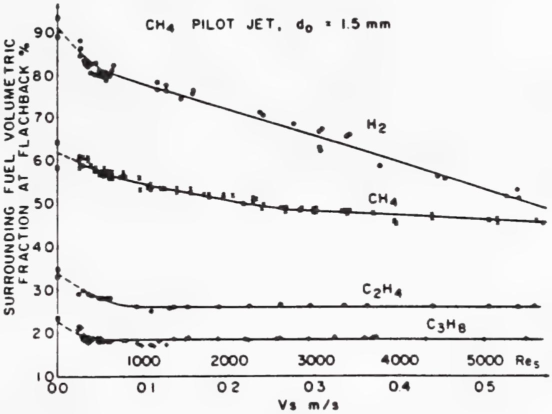

Fig. 15 Variation of the fuel volume fraction in the stream at flame flashback for various fuels vs. the surrounding stream velocity.

FLAME SPREAD OR FLASHBACK LIMITS

The lean flashback limits of mixtures with air of such single gaseous fuels as hydrogen, methane, ethylene, and propane were determined for varying stream velocities. As can be seen from Fig. (15) the flame flashback limits of ethylene and propane are little affected by the stream velocity, while for methane and hydrogen, they were affected significantly over the range considered. The exact reason for the significant decrease of the flame flashback limit of hydrogen and methane and to some extent, for ethylene and propane with increasing gas velocity is uncertain. However, it can be suggested that as the stream velocity is increased, the turbulence level in the flame zone increases, resulting in a higher flame velocity. Hydrogen and to a much lesser extent, methane, being relatively diffusive and light, experience a higher increase in flame speed, thereby enabling the flame to propagate at smaller concentrations.

It is also noted that the flashback limits of all the fuels under consideration are slightly lower than their corresponding lower flammability limits for downwards flame propagation under quiescent mixture conditions. This is basically due to two reasons. Firstly, the V-shaped flame front, before flashing back, attains a fairly large surface area that radiates heat into the surroundings. Therefore, the mixture ahead of the flame front becomes preheated as it approaches the flame zone, thereby allowing flame propagation at a slightly smaller concentration than the published lean limits, which are determined normally at room temperatures. Secondly, as the gas stream reaches the flame zone, it induces some turbulence in the interphase between the flame and the oncoming stream, which is further amplified by the self induced turbulence of the flame itself. The flame generated turbulence on the flame front is principally due to local thermal expansion, acceleration of burned products and the bending of the stream lines as the gas passes through the flame front. It is well recognized that an increase in turbulence level produces a wrinkled flame which in turn enhances the reaction rate as well as the heat and mass exchange between the flame and the oncoming stream. Moreover, the enhanced turbulence level results, up to a point, in a higher flame velocity (12,13). The combined effect of these factors allows the flame to propagate into the gas stream at a slightly lower concentration than normally expected (14).

The flashback limits of binary mixtures of the fuels under consideration were determined at a given stream velocity. Some typical results are shown in Fig. (16) where it can be seen that the flashback limit for a fuel mixture in air falls throughout somewhere between the corresponding limits for the fuel components making up the mixture in air. In these tests, the fuels were first mixed together in various proportions before being introduced into the air stream. The gas velocity was kept constant for all the tests. The main objective was to establish how the lean flashback limit for a binary fuel mixture in air is related to the corresponding limits for the individual fuels on their own in air under similar conditions. Moreover, it was necessary to find out whether predictive measures can be employed to establish the limits for fuel mixtures in the air.

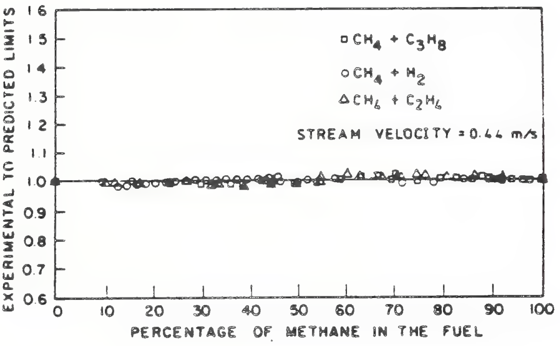

Fig. 16 Typical variation of the ratios of the experimental to the predicted flashback limits according to Le Chatelier's Rule in air for various binary mixtures of hydrogen, methane, ethylene and propane.

The flashback limit for a fuel-air stream as established here is obviously related to the flammability limit value under similar conditions. Accordingly, it would be expected that Le Chatelier's Mixing Rule normally employed for the determination of the lower flammability limit of some fuel mixtures in air can be considered for this case also. This Rule, which is based on the assumption that the mixture of limiting mixtures, is itself a limiting mixture, leads to the following relationship:

$$L_m = (\sum_N y_i/L_i)^{-1}$$

where y_i is the molar fractional component of item 'i' in the mixture, L_i is the flammability limit of the fuel 'i' on its own in air by volume and N is the number of fuel components making up the fuel mixture. Therefore, the experimentally determined flashback limits of the individual fuels under identical flow conditions were used for the theoretical predictions. It was found that Le Chatelier's Rule can be employed to predict the flashback limits of binary mixtures of gaseous fuels in air to within ±5% accuracy when the corresponding limits of the individual fuels under identical operating conditions are known. The nature of the scatter of the points suggests that much of the deviation was of random nature and could possibly be mainly influenced by experimental errors.

SOME MAJOR CONCLUSIONS

A brief statement of some of the main conclusions derived from this investigation follows:

i) The presence of some fuel in the surroundings of a fuel jet can extend its blowout limit. The combustion zone though will be confined to the pilot flame region and will be extended very significantly both in length and width as the fuel concentration in the jet surroundings is increased. Larger size pilot flames will correspondingly produce greater conversion of the fuel in the surroundings. Moreover, the temperature and concentration fields of the jet flame will be modified.

ii) Flame spread from the pilot flame into the surrounding lean mixture takes place once the fuel concentration in the surroundings reaches a critical concentration. For very small stream velocities, as the concentration of the fuel in the surroundings is increased, the flame propagation tends initially to be patchy and downstream of the jet. Somewhat higher fuel concentrations are needed to have the flame propagate or flashback against the surrounding stream. At higher stream velocities these two types of limits coincide to result in the flame propagating suddenly and against the surrounding stream.

iii) The flame flashback limits of gaseous fuels in air through a stream of homogeneously lean fuel-air mixtures in the presence of a continuously burning small pilot flame were lower than the published lean flammability limits (by about 25%) for downward propagation of the corresponding fuels for quiescent mixtures at atmospheric pressure and intake temperatures, but they tend towards the published values as velocities of the surrounding stream approach zero.

iv) The burning velocity for flame flashback conditions of the lean gaseous mixtures of the light hydrocarbons under consideration tends to be practically independent of the type of fuel for surrounding stream velocities below 60 mm/s. Moreover, at the end of the flame zone, the efficiency of the combustion of the very lean mixtures tends to be high and probably above 90%.

v) The flashback limits of binary mixtures of gaseous fuels can be predicted reasonably well by using Le Chatelier's Mixing Rule provided the experimentally determined flashback limits of the individual fuels under identical flow conditions are used in the relationship.

ACKNOWLEDGEMENT

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Flame Flashback for Low Reynolds Number Flows

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Abstract

The flame flashback in coflowing homogeneously mixed airfuel stream has been studied for a range of stream Reynolds number up to 6000. Methane, propane, ethylene, and hydrogen were employed as fuels in the surrounding stream. The flame lean flashback limits of gaseous fuels in the presence of continuously burning small pilot flame were lower than the published lean flammability limits for downward propagation of the corresponding fuels for quiescent mixtures at atmospheric pressure and intake temperature, but they tend to that value for velocities of the surrounding stream tending toward zero. The velocities of propagating flame at flashback are calculated on the basis of the critical angles of the leading edge of the flame front and an idealized model of the flame flashback shape. The combustion efficiency of the system for flow conditions near the flame flashback limits has been established.

Nomenclature

dc = diameter of the combustor, mm
do = diameter of the pilot flame inlet nozzle, mm
h = height of the measurement cross section above exit plane of the pilot flame, m
FVF = fuel volume fraction, %
HCO, HCH_4, HH_2, H_F = net calorific value of CO, unburned CH_4, H_2, and a fuel (CH_4), respectively, MJ/kg
EICO, $EICH_4$, EIH_2 = emission index of CO, unburned CH_4 and H_2, respectively, g/kg
LFB = lean flame flashback limit, %

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Lo = air-fuel stoichiometric ratio
M_A, M_F = molecular mass of air and fuel, respectively, kg/mol
$[O_2]$,$[CO_2]$,$[CO]$,$[CH_4]$,$[H_2]$,$[C_2H_4]$,$[C_3H_8]$ = volume fractions of O_2, CO_2, CH_4, H_2, C_2H_4, C_3H_8, respectively, %
r = radial distance, m
R = radius of the combustor, m
Res = Reynolds number of the surrounding stream
Vb = burning velocity, m/s
Vs = velocity of the surrounding mixture, m/s
β = angle of the flame front
ϕ = equivalent ratio
ηc = combustion efficiency

Introduction

The combustion of fuel in a nonquiescent homogeneous air-fuel mixture is very often encountered in many industrial combustion devices, including premixed prevaporizing combustors. Furthermore, it has recently been reported that the stability of the diffusion jet flame can be improved significantly by the surrounding flame from the homogeneously premixed coflowing air-fuel stream instead of air alone [Karim and Hanna (1984)][5].

One of the limitations of such an improvement of the main flame stability is the possibility of the flame flashback in the surrounding stream. Studies of the flame flashback for the open flames have been presented in detail by many investigators [e.g. Lewis and von Elbe (1951)[7]; Wohl et al (1949)[9]]. The present laboratory investigations of the flame flashback were carried out for the flow inside the combustor and were initiated as a continuation of the study by Karim et al (1985)[6].

The main objectives of the present work are as follows:

1) To investigate the flame lean flashback limit of the surrounding homogeneously mixed air-fuel stream, in the presence of the small pilot flame, mainly for the stream velocity range of less than 0.3 m/s, /Re $\leq$ 3000/.

2) To determine the burning velocity of the lean gaseous homogeneous air-fuel mixtures under flashback conditions.

3) To determine experimentally the combustion efficiency of the flame in the presence of the pilot flame for conditions of the flow near the flame lean flashback limit.

Experimental Setup

The experimental setup employed in this study was described in the previous works in detail [Karim et al (1981)[4]; (1985)[4]. The apparatus (Fig. 1) was a vertical steel combustor of 150mm diameter and 400mm height, open at the top and fitted with a special glass window for viewing the flame. A sharp-edged nozzle of 1.5mm diameter (Fig. 1b), located along the centreline of the combustor, was used to produce as small a diffusion pilot flame as possible, which acted as the ignition source for the surrounding premixed fuel-air stream. The surrounding stream flows from the bottom of the combustor. Before entering the combustor, the flow passes through a settling chamber, a perforated cone, a set of stainless steel wire mesh, and a honeycomb flow straightener for producing a possibly uniform velocity distribution at the inlet section of the combustion chamber. The wire mesh also acts as a flame arrestor in case of flashback through the honeycomb. The central jet diffusion flame is ignited initially by an external ignition source, which is later removed.

Various fuels are supplied from a high-pressure cylinder, while air·is taken from the compressed air supply of the laboratory. The fuel used in the diffusion pilot flame was commercial methane, typically composed of 94.7% CH_4, 3.7% C_2H_6, 0.7% C_3H_8, 0.2% C_4H_{10}, 0.4% C_5H_{12}, and 0.3% N_2. The surrounding fuels involved were methane, propane, ethylene, and hydrogen. The surrounding stream velocity ranged from 0.025 to 0.60 m/s. The volume flow rates of the fuels and air were measured using choked nozzles, and the profile

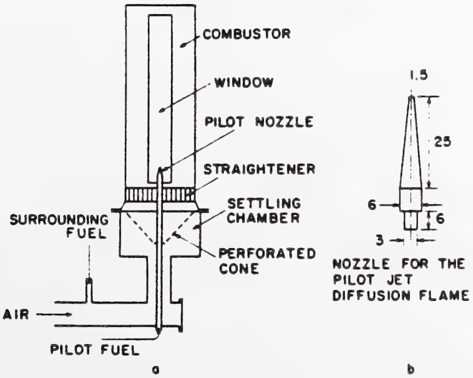

Fig. 1 Schematic diagram of the experimental setup.

of the velocity of the surrounding stream was measured by means of a hot-wire anemometer probe. The analysis of the combustion gases was carried out by means of Beckman analyzers that used a water-cooled stainless steel 2.5mm diameter probe for sampling.

The angle of the flame front at the moment of flashback was measured using the video technique. The fuel concentration in the surrounding stream was slowly increased up to the flashback conditions, and the pictures of the flame shape were recorded on videocassette using a HITACHI HV9017. The visible angle of the flame front was later measured from the TV screen.

There are three possible approaches to the determination of the flame flashback of air-fuel mixtures:

1) The concentration of a fuel in the stream is kept constant, and the flame flashback is obtained by varying the stream velocity.
2) The velocity of the stream or air is kept constant, and the flame flashback is obtained by varying (increasing) the fuel concentration. With an increase in the fuel concentration in the stream velocity, this change is small and easy to calculate.
3) The flow rate of the fuel is kept constant, and the flame flashback is obtained by varying (mainly decreasing) the stream velocity. This approach provides varying velocities of the stream and fuel concentrations.

Because the first approach was impossible to perform for this setup, the second and third were used. Thus, the limits obtained as a result of this investigation are the lean limits for flame flashback.

Results and Discussion

The pilot fuel flow rate was very small and constant for all ranges of this investigation. As a fuel was introduced into the surrounding stream, combustion initiated along the periphery of the pilot flame, even at very small concentration. Practically, it was impossible to find any threshold of the concentration for which the combustion at the vicinity of the pilot flame does not exist.

Figure 2 shows the variations of the pilot flame with increasing fuel concentration in the surrounding stream. In the case of FVF = 0 (Fig. 2a), the pilot flame zone is a typical zone of an overventilated jet diffusion flame for a small Reynolds number (Re = 20). The pilot zone is surrounded by a strong, easily visible, light-blue combustion zone about 0.5 mm thick and a very weak light-blue

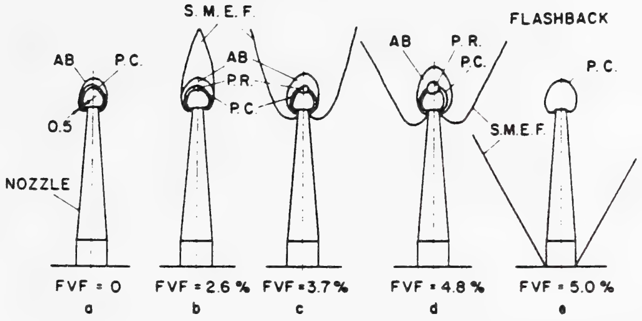

Fig. 2 Variation of the pilot flame zone; Vs = 0.23 m/s, scale 2:1, do = 1.5mm, CH_4 pilot fuel, Rej 20; AB, after burner zone, P.C. pilot combustion zone; SMFF, surrounding mixture flame front; P.R., pilot combustion rich zone.

afterburning region. As fuel is introduced into the surrounding stream, a new zone appeared around the pilot flame (Fig. 2b). The pilot zone scarcely varies its appearance under these new conditions. This is probably because enough oxygen exists after the surrounding mixture flame zone. With a further increase in the fuel concentration of the surrounding stream, the afterburning region is increased, and the top of the combustion zone (Fig. 2b-d) becomes a visible rich yellow-orange. This rich combustion zone also grows as the concentration of the fuel is increased. Figure 2d shows the pilot flame zone just before the flame flashback. The rich combustion zone, the strong combustion zone, and the afterburner region disappear after flame flashback (Fig. 2e).

Figure 3 shows the schematic representation of the flame propagation from the pilot flame into the surrounding stream. With an increase in the concentration of the fuel, the surrounding mixture flame zone became wider and higher. The tip of the flame became opened up for some concentration of the fuel (Fig. 3j), and V-shape flames are produced. The angle of the V-shaped flame continuously increased with increasing concentration until it reached a critical angle termed "flashback angle" (Fig. 3l). Increasing the concentration beyond this point caused the flame to flashback on the base of the combustor.

The lean flashback limits of mixtures with air of such single gaseous fuels as hydrogen, methane, ethylene, and propane were determined for varying stream velocities, and

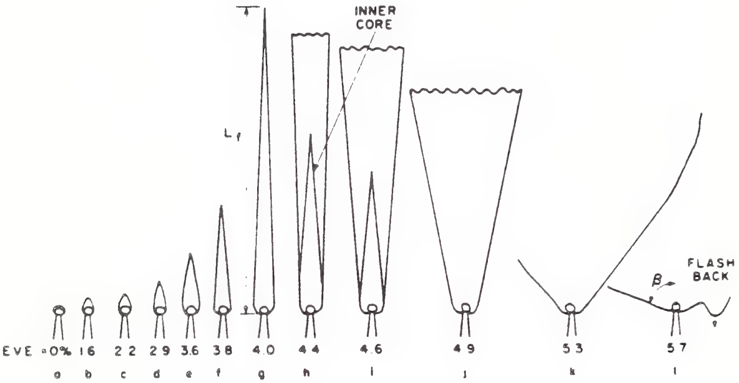

Fig. 3 Schematic representation of the flame propagation from the pilot flame into the surrounding stream; Vs = 0.06 m/s, CH_4 pilot fuel, CH_4 surrounding fuel, do = 1.5mm.

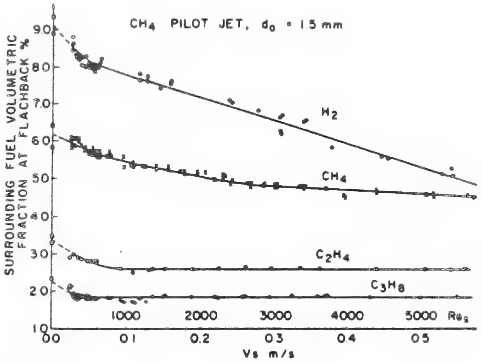

Fig. 4 Variation of the fuel volume fraction in the stream at flame flashback for various fuels vs. the surrounding stream velocity.

the results are shown in Figs. 4 and 5. It is observed that the lean flame flashback limits over the range of stream velocity considered are lower than the lean flammability limits for downward flame propagation in quiescent mixtures of the corresponding fuels [see Coward and Jones (1952)[3]; Burgess and Hertzberg (1975)[1]]. The flame flashback limits of ethylene and propane are only in the narrow range affected by the stream velocity over the range considered. Beyond these ranges of influence of Reynolds number (Res = 800 for air-ethylene mixture and Res = 460 for air-propane mixture), the lean flame flashback limits were independent of the velocity of the surrounding stream and equal to 75% of its lean flammability limits for downward flame propagation.

The flame flashback limits of methane and hydrogen were affected by the stream velocity for the entire range considered. The exact reason for the significant decrease of the flame flashback limit of hydrogen and methane and, to some extent, for ethylene and propane with increasing gas velocity is uncertain. Some explanation of this phenomenon was given by Karim et al (1985)[6].

Figure 5 shows the variations of the equivalence ratio of the surrounding stream at the flame flashback conditions for fuels under consideration. The curves $\emptyset = f(Vs)$ are

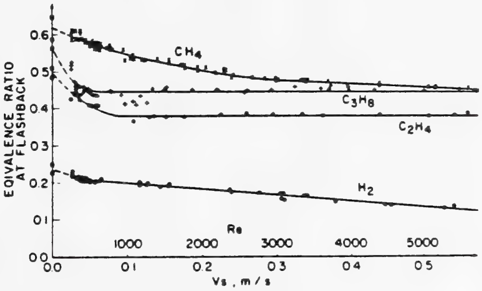

Fig. 5 Variation of the equivalence ratio of the stream at flame flashback conditions for various fuels vs. the surrounding stream velocity; CH_4 flame pilot jet, do = 1.5mm.

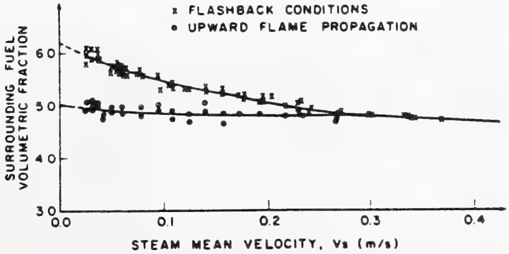

Fig. 6 Comparison of the variation of the fuel volume fraction in the stream at flame flashback and at flame upward propagation for CH_4 vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5mm.

arranged in another order in view of the following relationship:

$$\emptyset = \frac{LFB \cdot Lo}{(MA/MF) \cdot (100-LFB)}$$

It is observed that for some concentrations of the fuel, just before flame flashback started, large portions of the flame tore off from the main surrounding flame. It seems to be quite similar to the lean flame upward propagation. Figure 6 shows the lean limit of this flame upward propagation in comparison to the lean flame flashback limits, both for CH_4 fuel.

The flashback of the flame did not occur at the same time over the entire area of the combustor. It should be noted that for the surrounding stream velocity $Vs > 0.15$ m/s ($Res = 1500$), as concentration of fuel in the stream was increased gradually, the flame first started flashing back along the pilot flame nozzle. The angle of the flame front was constant during this flashing back (Fig. 7a). The flashback of the flame for the surrounding stream velocity $Vs < 0.1$ m/s started first far from the pilot flame nozzle (Fig. 7b,c). The flashback of the flame for the surrounding stream $0.15 > Vs > 0.1$ m/s started in a way similar to the case of Fig. 7a or of Fig. 7b and c. The flashback of the flame for the surrounding stream velocities $Vs < 0.05$ m/s, and especially for heavier hydrocarbons (C_3H_8), proceeded in a considerably longer time. Flashback usually occurred after the flame position began oscillating.

For the constant pressure and temperature of the surrounding stream, the flame flashback is dependent on the

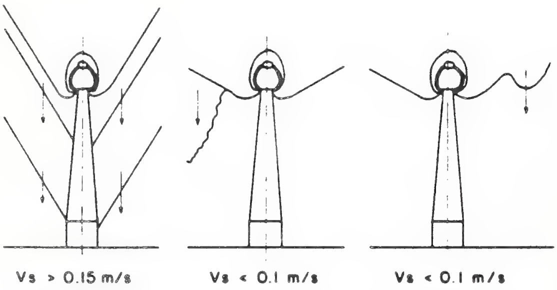

Fig. 7 Schematic representation of the flame flashback a) flame flashback for $Vs > 0.15$ m/s; b,c) flame flashback for $Vs < 0.1$ m/s.

local concentration of the fuel and the local velocity. The measurements of the local concentration of the methane vs. radial distance of the combustor are shown in Fig. 8. It can be seen from this diagram that the profiles of the concentrations of CH_4 are uniform. Figure 9 shows the profiles of the surrounding stream velocity for the flow conditions Vs = 0.025, 0.058, and 0.22 m/s without the pilot fuel flow, with the pilot fuel flow and with the pilot flame.

It is seen from these diagrams that the pilot fuel jet drastically changed the profile velocity in the vicinity of the nozzle (for a radius of up to 10mm). For the fuel pilot jet with pilot flame, these changes are not so strong, however, and probably for a flow with surrounding flame, these profiles of velocity are more equalized. It seems that for Vs < 0.1 m/s for the flow with a surrounding flame, the local velocities in the vicinity of the pilot flame are less than the mean velocity (Vs). We had no opportunity to measure the local velocities of the surrounding mixtures in the vicinity of the pilot flame for flows with the surrounding flame. From these considerations, we can draw a conclusion that for a homogeneous surrounding stream and an equalized profile of the velocity, the flame flashes down at first in the vicinity of the pilot flame, where the thermal conditions for the combustion of fuel-air mixtures are better.

It was observed that the critical angle between the leading edge of the flame front and the vertical axis of the combustor at which the flame flashes back was different for a different fuel. This angle was the smallest for hydrogen and the largest for propane. For the same fuel, an in-

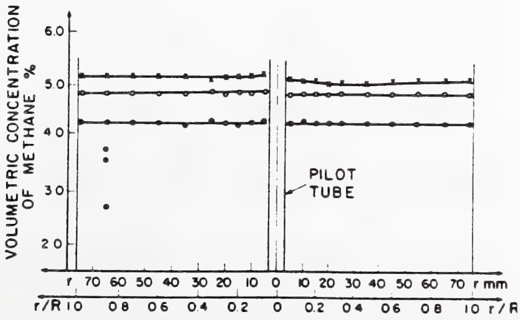

Fig. 8 Measured fuel volume fraction of CH_4 in the surrounding stream vs. radial distance of the combustor; Vs = 0.06 m/s, h = -18mm, CH_4 flame pilot jet, do = 1.5mm.

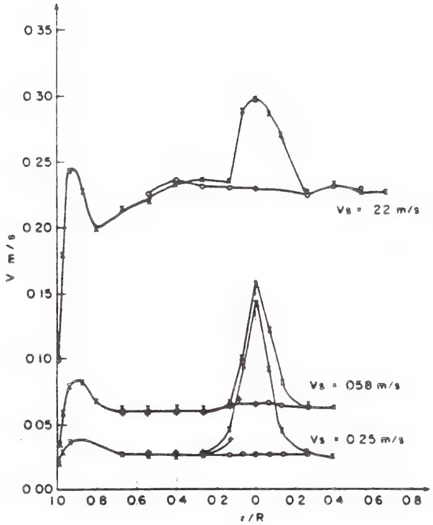

Fig. 9 Comparison of the profiles of the velocities for flow conditions with the pilot fuel flow (—x—), without the pilot flow (—o—), and with the pilot flame (—+—); CH_4 pilot fuel; do = 1.5mm, Rej #20.

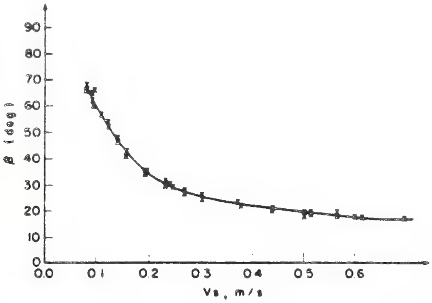

Fig. 10 Variation of the front flame angle at the beginning of flame flashback for CH_4 vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5mm.

crease in surrounding velocity caused a decrease in the critical angle.

Figures 10 and 11 show the critical angle of the flame front for methane, ethylene, and propane. For light hydrocarbon fuels and surrounding velocities Vs > 0.15 m/s, this angle does not vary during the flame flashback (Fig. 7a),

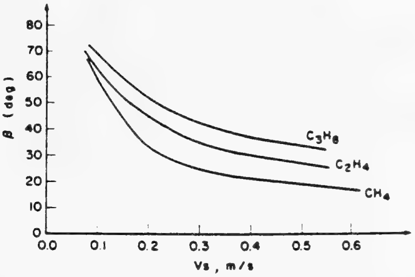

Fig. 11 Comparison of the variation of the front flame angle at the beginning of flame flashback for various hydrocarbon fuels vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5mm.

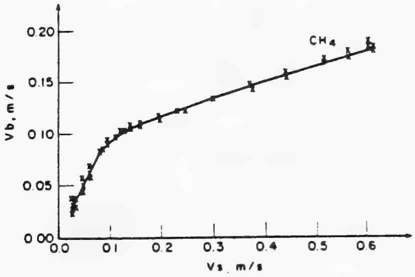

Fig. 12 Variation of the burning velocity at the beginning of flame flashback for CH_4 vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5mm.

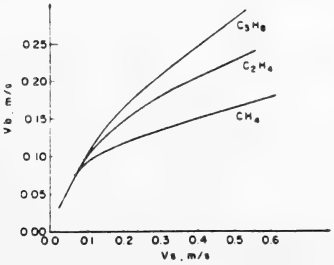

Fig. 13 Comparison of the variation of the burning velocity at the beginning of flame flashback for various hydrocarbon fuels vs. the surrounding stream velocity; CH_4 pilot jet, do = 1.5mm.

and it is the critical flame flashback angle. For velocities Vs < 0.15 m/s, the critical angle could be varied during the flame flashback and, for that case, it is the angle just before flame flashback. For velocities Vs < 0.08 m/s, this angle is very difficult to determine. Some attempts to measure the critical angle for hydrogen by using the same method as for hydrocarbon (that is, without intervention in the flow) failed because of the dimness of the hydrogen flame for this concentration.

It can be seen from these diagrams that for a velocity Vs = 0.55 m/s, the critical angle for propane is about 75% greater than for methane and about 25% greater than for ethylene. For small velocities Vs < 0.1 m/s, this difference between the critical angles for different fuels is comparatively small.

The burning velocity is calculated for an idealized model of the flashback flame shape. It is assumed that the surface of the flame forms an idealized cone without any waving of its surface (with no wrinkled flame). With these assumptions, the burning velocity can be calculated from the expression

$$V_b = V_s \cdot \sin\beta$$

Figures 12 and 13 show the variation in the burning velocity calculated from the preceding expression. It can be seen from the diagram (Fig. 13) that, for the mean surrounding velocity Vs = 0.55 m/s, the burning velocity of propane is about 70% greater than that of methane and about 22% greater than that of ethylene. For the surrounding velocity Vs < 0.1, these differences between the burning velocities are comparatively small. For surrounding velocities Vs < 0.06, there is virtually only one burning velocity for those three hydrocarbons.

Gas analyses were carried out to determine the radial and axial distributions of the concentration of methane before the combustion zone and of carbon dioxide, carbon monoxide, oxygen, and unburned CH_4 for the combustion zone. Knowledge of these concentrations enables us to calculate the combustion efficiency and the equivalence ratio for the measurement points.

Gas analyses for the combustion zone (cone of the flame) could not be performed for a fuel concentration in the surrounding stream very near the lean flame flashback limit because the probe and the gas sample caused the strong local perturbation of the flow, and the flame started flashing back much earlier than it would have without mechanical perturbation. For these reasons, gas samples were taken for analysis for three concentrations of methane: 4.25, 4.8, and 5.1 - 5.3% step by step closer to the lean flame flashback limit (LFB = 5.7%). The gas analyses were carried out for five cross sections along the flame. The nearest axial location to the exit plane of the nozzle was for a height h = 7mm, just at the end of the main pilot flame. The second location was for h = 12mm, just above the afterburner pilot zone. Then there were two intermediate stations for h = 22 and 57mm. The farther station was located for h = 97mm, where the combustion reactions ought to have been finished. Gas analyses were carried out for only one mean velocity of the surrounding stream Vs = 0.06 m/s (Res = 600).

The adiabatic efficiency of combustion is calculated from the expression:

$$\eta_c = 1 - \frac{(EICO \times HCO + EICH_4 \times HCH_4 + EIH_2 \times HH_2) \times 10^{-3}}{H_F}$$

We had no opportunity to measure the concentration of H_2 in the flame during our experiments. However, bearing in mind that, according to Clifford et al (1977)[2], the concentration of unburned H_2 in the combustion gases at the outlet is less than one-tenth that of CO and that the oxidation rate of H_2 under the conditions of interest are about one order of magnitude higher than that of CO, we assumed that the concentration of H_2 was $[H_2] = 0$. With

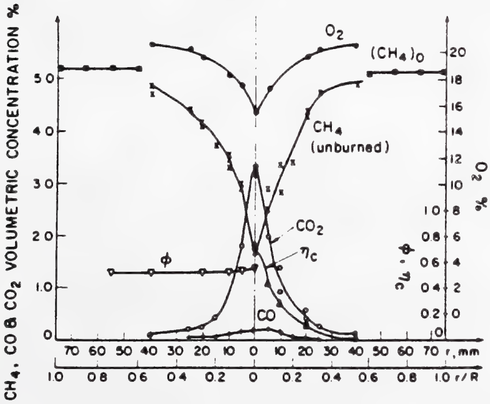

Fig. 14 Volume fractions of unburned CH_4, CO, CO_2, and O_2 vs. radial distance; initial volume fraction of CH_4 in the surrounding stream (CH_4) = 5.1%, Vs = 0.06 m/s, CH_4 pilot flame, do = 1.5mm, h = 22mm.

such a limitation after transformation, and introducing the values of MCO, MCH_4, HCO, and $H_F = HCH_4$ [see Rose and Cooper (1977)]^8, we have

$$\eta_c = \frac{[CO_2] + Bo \times [CO]}{[CO_2] + [CO] + [CH_4]}$$

The equivalence ratio of the combustion gases is calculated, after similar transformation, from the following expression:

$$\phi = \frac{B_1 \times ([CO_2] + [CO] + [CH_4])}{100 - [CO_2] - 1.5 \times [CO] - [CH_4]}$$

where, for methane as a fuel, Bo = 0.6474 and B_1 = 9.515.

Figure 14 shows the concentration profiles of CH_4, O_2, CO_2, CO, and unburned CH_4 for the station h = 22mm above the nozzle for a combustible mixture with 5.1% methane. These measurements were carried out for all the radial distances, from side to side, together with measurements of initial concentration of methane.

Figure 15a-d show the radial distribution of unburned CH_4, CO_2, CO, and O_2 for the four stations and for the possibly higher initial concentration of methane (5.35 - 5.3% CH_4). Figure 16 shows the radial distributions of unburned CH_4, CO_2, CO, and O_2 and the calculated efficiency of combustion η_c for the farthest station (h = 97mm) and

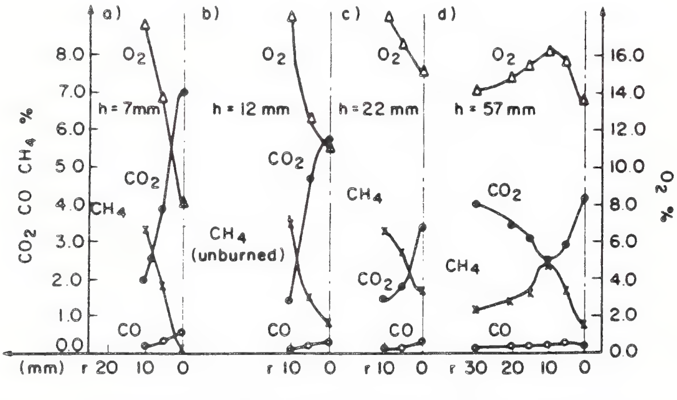

Fig. 15 Volume fractions of unburned CH_4, CO, CO_2, and O_2 vs. radial distance for few measurement heights (h = 7, 12, 22, 57mm); initial volume fraction of CH_4 in the surrounding stream $(CH_4)_o$ = 5.3-5.25%, Vs = 0.06 m/s, CH_4 pilot flame, do = 1.5mm.

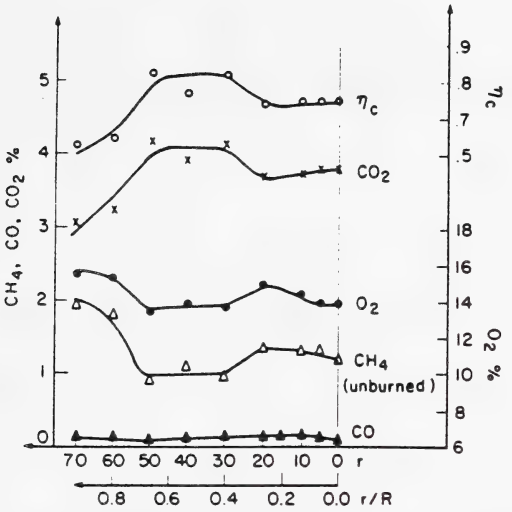

Fig. 16 Volume fractions of unburned CH_4, CO, CO_2, O_2 and combustion efficiency (η_c) vs. radial distance for h = 97mm, $(CH_4)_o$ = 5.15%, Vs = 0.06 m/s.

for a possibly highest concentration of methane. It can be seen from the diagram that for this station the distribution of the concentrations of the gases under consideration are more equalized. From these results, we can draw the conclusion that, for the critical initial concentration of methane, the concentration profiles of the combustion gases will be more equalized and the combustion efficiency much higher.

Conclusions

We have experimentally determined the flame lean flashback limits, the angles of the flame front, the concentrations of CO_2, CO, unburned CH_4, and O_2 in the cone of the flame, and the initial concentration of the methane in a nonquiescent stream of premixed fuel and air. The burning velocity, combustion efficiency, and equivalence ratio were calculated on that basis. Those investigations led to the following conclusions:

1) The flame lean flashback limits of various individual gaseous fuels in a stream of almost homogeneously premixed fuel and air within the range of velocities Vs =

0.25 to 0.6 m/s are in good agreement with previous experiments by Karim et al (1985)[6].

2) The flame flashback limits of gaseous fuels in air through a stream of homogeneous lean fuel-air mixtures in the presence of a continuously burning small pilot flame were about 25% lower than the published lean flammability limits for downward propagation of the corresponding fuels for quiescent mixtures at atmospheric pressure and intake temperature, but they tend toward published values as velocities of the surrounding stream approach zero.
3) For the surrounding homogeneous stream and equalized profile of the stream velocity, the flame flashing down at first in the vicinity of the pilot flame.
4) A V-shape flame front is formed for a velocity of the surrounding stream higher than 0.1 m/s. The angle between the leading edge of the flame front and the vertical axis of the combustor for the flame flashback conditions can be used to calculated the mean burning velocity for lean air-fuel mixtures.
5) For velocities of the surrounding streams Vs > 0.1 m/s, the burning velocity of the lean mixtures of the light hydrocarbons under consideration depends on the kind of fuels and the stream velocity.
6) The burning velocity of the lean gaseous mixtures of the light hydrocarbons under consideration is practically independent of the kinds of fuel for surrounding stream velocities below 0.06 m/s.
7) The combustion of the fuel is not finished in the visible flame front but is continued along the cone of the flame.
8) For the flashback conditions of flow, at the end of the flame zone, the efficiency of combustion is very high, probably above 90%.

Acknowledgment

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PUBLICATION NO. 5

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Examination of the Formation of Combustible Mixtures by Diffusional Mass Transfer

The approximate prediction of the concentration profiles following the release of a fuel into air, typically within a confined space at constant pressure and temperature is presented using generalized charts based on the one-dimensional diffusion equation and an effective eddy diffusivity. It is suggested that such generalized plots can be employed to establish the size and changes with time of the associated flammable zones, including when convective diffusion may be involved. Examples for some typical common fuels are presented.

Introduction

The stratification of combustible mixtures at constant pressure and temperature is encountered in numerous hazardous situations and in many technical applications. Leakage of fuels from storage tanks or pipelines, oil or gas spills during transportation, formation of layered combustible mixtures within rooms, corridors, elevator shafts, mine galleries and tunnels and the formation of stratified mixtures in gas turbines, furnaces, and internal combustion engines are some examples. Such mixtures are mostly formed by mass transfer phenomena that remain inadequately understood. Further research is needed so as to improve the safety guidelines relating to such hazardous situations.

Early work on the formation of stratified combustible mixtures was directed towards examining fire hazards, especially in mine galleries (Coward and Georgeson, 1941; Liebman, Corry and Perlee, 1971). The formation and dispersion of methane along roof layers due to buoyancy effects were studied (Bakke and Leach, 1962) and the ventilation rates required to reduce the fire hazards were determined (James and Purdy, 1962). Factors affecting buoyancy-driven flows were also studied (Turner, 1973; Karim and Tsang, 1975; and Badr, 1985) and one-dimensional models for convective diffusion were developed and checked against experiment. Karim et al. (Karim and Lam, 1986; Karim, Kibrya, Lam, Petela and Wierzba, 1987; and Karim, Lam, Petela, and Rowe, 1987) studied the formation of combustible mixtures following the discharge of gaseous fuels into air and the fuel concentration profiles were measured. They suggested that the simple one-dimensional molecular diffusion equation may be usable to reproduce such data while replacing the molecular diffusivity by a variable much larger effective diffusivity.

The present contribution also considers the approximate prediction of the fuel concentration profiles and the associated flammable zones following the leak or release of a fuel typically into a confined space of air at constant pressure and temperature.

In many practical situations, similar to those discussed earlier, when a gaseous fuel leaks into a vertical confined space of air, the formation of two gas layers with an interface between them may result. If the upper fluid is lighter than the lower one, the two fluids are stable and mixing occurs by molecular diffusion. However, for ideal fluids, if the upper fluid is denser, the fluids are unconditionally unstable, leading to convective diffusion. For real fluids, molecular viscosity and diffusion can act to damp out some of the disturbances. Thus, a criterion needs to be developed to establish whether the fluid layers are stable or unstable. Although such common situations are obviously very important from a safety point of view, no such reliable formulation is yet available. Much of the previous work on the phenomenon of convective diffusion within a confined space was very limited to the approximate prediction of fuel concentration profiles for specific conditions without getting into the details of the ensuing flow patterns and their stability criteria. However, some conclusions can be arrived at by analogy with the similar problem of thermal convection such as in a horizontal fluid layer heated from below. The similarity between the two problems stems from the fact that both have positive vertical density gradient as the driving force for the unstable convective flows.

The main conclusion that can be made in relation to the mixing relating to the situation being considered here, shown schematically in Fig. 1, where a heavier gas is overlying a lighter one within a long circular relatively large diameter tube, is that mixing by turbulent convective currents may be dominant even when the density difference is relatively small. However, when the mixing occurs in relatively small diameter tubes, the stabilizing effects of the side walls may cause molecular diffusion to dominate the mixing process. No effective quantitative guidelines, however, can be made to permit the calculation of concentration profile changes with time for a wide range of conditions.

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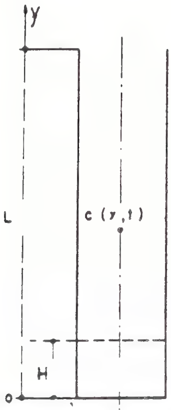


Fig. 1 Schematic representation of the fuel compartment with a height (H) and that for the air a height of ($L-H$)

Theory

In order to consider a theoretical model of the convective diffusion of a light fuel component vertically into air within the cylindrical tube of Fig. 1 at constant temperature and pressure, the fluid is imagined to be divided into a large number of infinitesimal stream tubes such that the velocities in any two adjacent tubes at the same vertical location (y) are equal in magnitude and opposite in direction. Hence, the heavier fluid flows downwards in one tube while the lighter fluid flows upward in an adjacent one. Accordingly, the governing equations associated with this simplified one-dimensional convective flow involving a binary mixture of fuel and air within the tube of Fig. 1 are the following:

$$\frac{\partial(\rho m)}{\partial t} + \operatorname{div}(\rho\, m\, \mathbf{W}) = \operatorname{div}(D \operatorname{grad} m) \tag{1}$$

for the mass conservation of the diffusing gas, and

$$\rho \frac{D\mathbf{W}}{Dt} = F - \operatorname{grad} P + \mu \nabla^2 \mathbf{W} + \frac{1}{3}\mu(\operatorname{div}\mathbf{W}) \tag{2}$$

for the conservation of momentum. m is the mass fraction of the diffusing gas, ρ is the density of the mixture, $\mathbf{W}$ is the velocity vector, F is the body force, and D is the binary diffusion coefficient. When laminar conditions, constant properties and Boussinesq approximation are assumed, these two equations reduce to the following forms:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial y^2} - \frac{\partial(CV)}{\partial y} \tag{3}$$

and

$$\rho\left(\frac{\partial V}{\partial t} + \frac{V\partial V}{\partial y}\right) = -\rho g - \frac{\partial P}{\partial y} + \frac{4}{3}\mu\frac{\partial^2 V}{\partial y^2} \tag{4}$$

where V is the velocity of one of the two fluids in the infinitesimal stream tube. These equations can be solved to obtain the velocity and concentration profiles as functions of diffusion time (Tsang, 1974; Badr, 1977). The solutions, however, are limited to small concentration gradients. For large concentration gradients such simplified solutions fail seriously to predict the concentration profiles since the relevant Rayleigh number becomes very high and turbulent unsteady three-dimensional convective currents dominate the flow field.

In situations dominated by turbulent convection, accurate prediction of the flow pattern and the distributions of concentration and velocity require the solution of the coupled multidimensional conservation equations (3) and (4) with the use of both appropriate eddy viscosity and eddy diffusivity values. Even when these requisite fluid properties are known, such solutions are extremely complicated, impractical and their ultimate accuracy is questionable. Unlike viscosity, reliable models for eddy diffusivity are unavailable in the literature. The application of suggested relationships such as that by Shirtcliffe between the turbulent diffusivity, turbulent thermal conductivity, Rayleigh number and the critical Rayleigh number for initiating convection to a situation such as the one being considered here is very questionable (Shirtcliffe, 1969). Other researchers (Uhl and Gray, 1966; Sherwood, Pigford and Wilke, 1975) showed that the eddy diffusivity is proportional to the mixing length of the eddies and the turbulent fluctuating velocity which are difficult to estimate.

The issue of the eddy diffusivity and the uncertainty of its available models led us (Karim and Lam, 1986; Karim, Kibrya, Lam, Petela, and Wierzba, 1987; Karim, Lam, Petela and Rowe, 1987) to suggest that under the circumstances, a much simpler approach for the prediction of the cross-sectional averaged fuel concentration profiles in tubes, similar to that shown in Fig. 1 may be employed. By using an appropriate effective diffusivity, the simple one-dimensional diffusion equation can then be employed. Such an effective diffusivity, expectedly, will be a function of many parameters, including the local concentration, concentration gradient, tube geometry, and properties of the two gases. We were able to reproduce the experimentally measured concentration profiles of methane diffusing upwards into air by using a variable effective diffusivity that is mainly a power function of the local concentration gradient. For example, an effective diffusivity of 0.005 m^2/s was used for short diffusion times while 0.0012 m^2/s for longer diffusion times at regions away from the initial interface of the two gases. This variation of the effective diffusivity with time may be attributed to the corresponding variation in the density gradient which in turn affected the intensity of turbulence and the mixing length.

Nomenclature

B = parameter describing tube geometry = L/H
C = volumetric concentration of diffusing fuel
C_{01} = initial concentration of fuel in lower section of tube
C_{02} = initial concentration of fuel in upper section of tube
D = coefficient of diffusion
F = body force
g = gravitational acceleration
H = length of fuel compartment
L = length of entire tube
m = mass fraction of diffusing fuel
n = number of terms in series solution
p = absolute pressure
t = time
V = velocity component in y direction
$\mathbf{W}$ = velocity vector
y = distance along tube axis
γ = dimensionless distance along tube axis = y/L
ρ = density of gas mixture
μ = dynamic viscosity of gas mixture
τ = dimensionless diffusion time = Dt/L^2
ξ = relative concentration = $(C-C_{02})/(C_{01}-C_{02})$

Subscripts

LFL = lower flammability limit
UFL = upper flammability limit

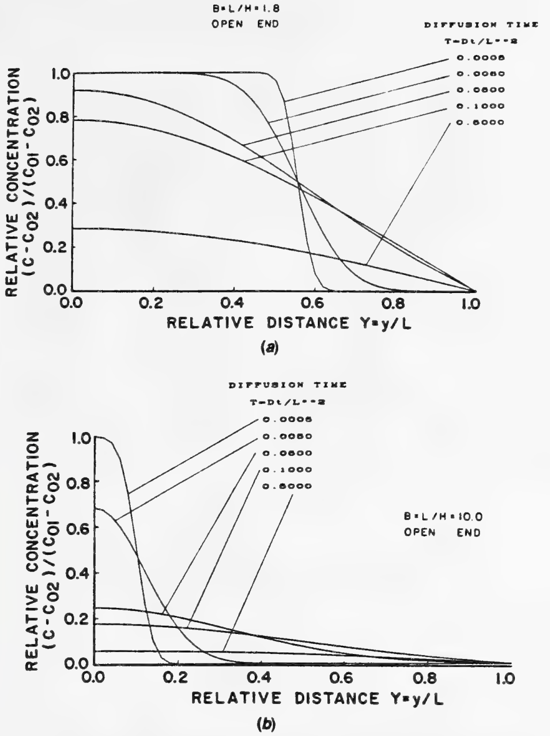


Fig. 2 Calculated distribution of the relative concentration of the diffusing fuel component in air with distance at constant temperature and pressure for a number of dimensionless times when the tube is having an open end on the air side

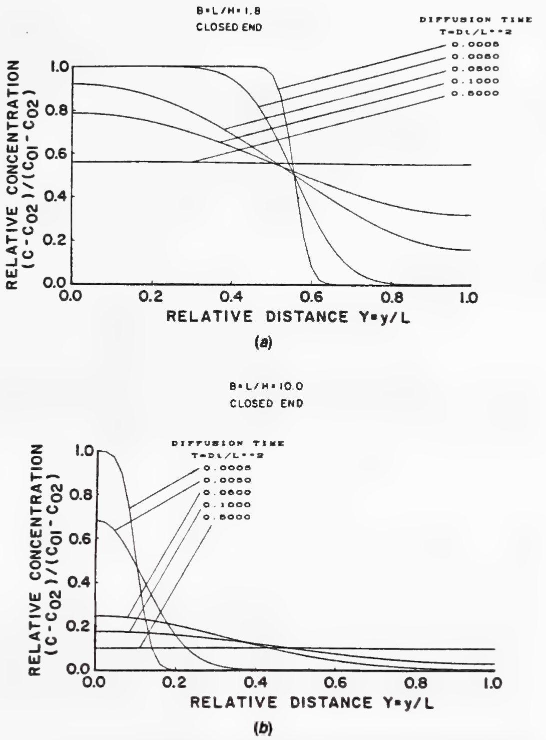


Fig. 3 Typical calculated distribution of the relative concentration of the diffusing fuel component in air with distance at constant temperature and pressure for a number of dimensionless times when the tube is having a closed end on the air side

The effective diffusivity approach appears much simpler and more practical than attempting to solve the corresponding conservation equations, especially when information about the flow patterns and velocities is not needed. Of course, more research is required to correlate the effective diffusivity to the parameters describing convective diffusion better. For the time being and on the basis of such an approach, however, generalized concentration profiles can be obtained and used for convective diffusion cases provided an adequate estimate of the effective diffusivity is made.

Concentration Profiles and Formation of Flammable Zones by Mass Transfer

Solutions to the simple one dimensional diffusion equation with constant properties are widely available in the literature (Jost, 1952; Jacobs, 1957). However, the use of a generalized dimensionless diffusion time τ being equal to Dt/L^2 in such calculations is advantageous particularly if it is going to be employed to consider the concentration profile under convective diffusion. Any of the generalized concentration profiles calculated on this basis and shown typically in Fig. 2 can then be considered to have been obtained either after a long diffusion time when mixing occurs by molecular diffusion or a much shorter time when mixing occurs by convective diffusion such that the value of $\tau = D_{\text{eff}} \cdot t/L^2$ stays the same.

Figures 2 and 3 show the concentration profiles for different boundary conditions, tube geometries, and diffusion times. The main feature of such profiles is that they can be used when mixing by molecular diffusion occurs between any pair of gases with any initial concentrations contained in tubes with different boundary condition, geometry, and inclination. To use such profiles, the values of the parameters C_{01}, C_{02}, h, L/D, and whether the tube end is open or closed are required. Moreover, as suggested previously, these profiles may also be usable when mixing between the two gases occurs by laminar or turbulent convection, provided an appropriate effective diffusivity is chosen and used.

It is of practical importance to consider the limiting case when a gas leaks from a pipeline or a duct into a relatively limited space of air; i.e., $B = L/H \rightarrow 1.0$. Another limiting case is when a gas leaks from a storage tank into a long tunnel or corridor; i.e., $B = L/H \rightarrow \infty$. Figure 4 shows the concentration profiles for such situations. It should be noted that when L/H is very large ($B \rightarrow \infty$), the boundary condition at the tube end expectedly has no effect on the solution.

One of the most important safety issues when a gaseous fuel leaks from a pipeline or a storage tank into a confined volume of air is the formation, growth, and decay of the flammable zone as a function of the time from initiating the mixing process. In the present study, this information can be made available by solving the inverse diffusion problem and establishing the contour lines of constant relative concentration (ξ) which becomes the same as the volumetric concentration (C). Knowing the type of fuel that is involved in the leak, its corresponding lean and rich flammability limits, its diffusivity in air (D), the overall dimensions of both the fuel and air tanks, (H) and (L), respectively, the size and the extent of growth of the flammable zone can be readily established. In using such profiles to find the size of the flammable zone after a given time from the start of the leak, either the appropriate molecular or the effective diffusivity could be used if mixing occurs by molecular or convective diffusion, respectively.

As an example of such an approach, the flammable zones for some common fuels are shown shaded in such typical plots

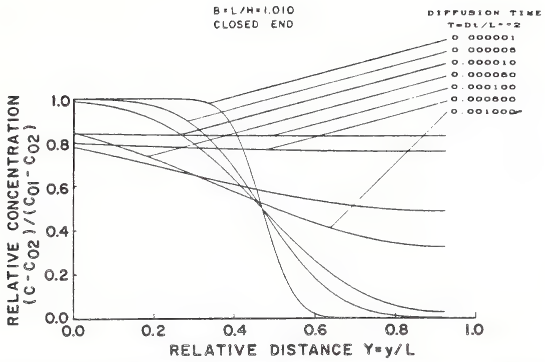


Fig. 4(*a*) Calculated distribution of the relative concentration of the diffusing fuel component into air for the special cases when $B = L/H$ is 1.01 when the tube is having a closed end for a range of dimensionless diffusion time

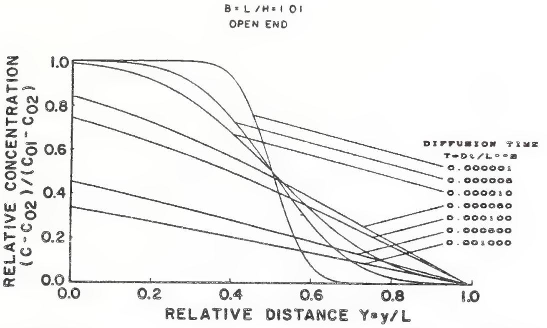


Fig. 4(*b*) Calculated distribution of the relative concentration of the diffusing fuel component into air for the special cases when $B = L/H$ is 1.01 when the tube is having an open end for a range of dimensionless diffusion time

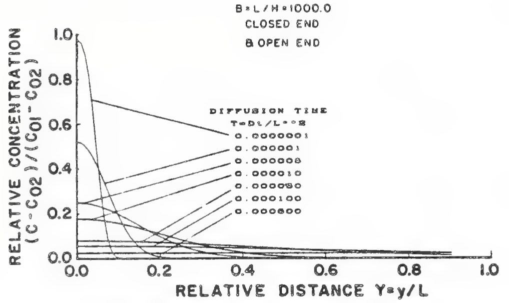
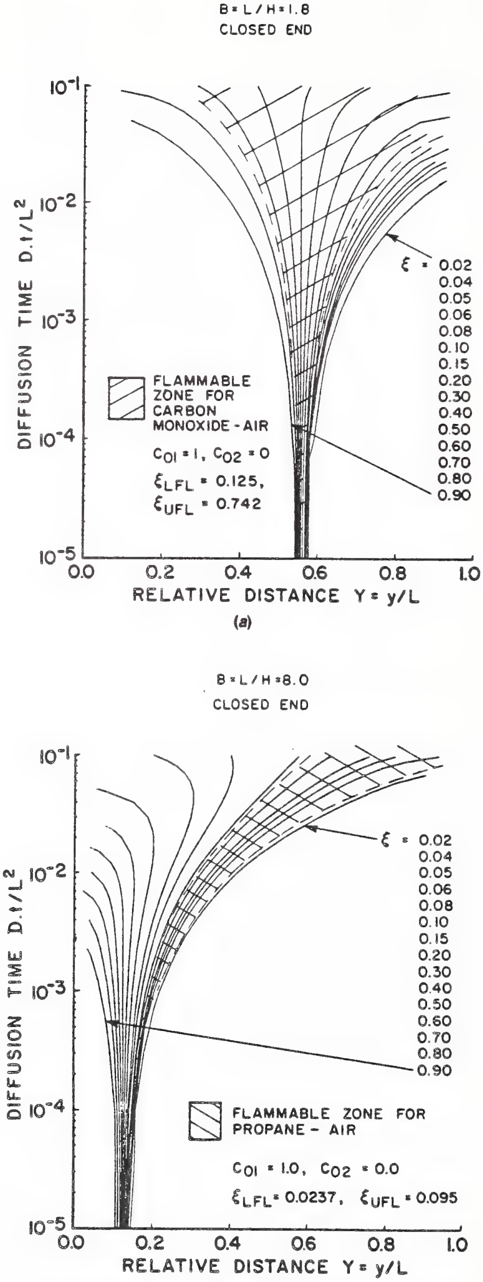


Fig. 4(*c*) Calculated distribution of the relative concentration of the diffusing fuel component into air for the special cases when $B = L/H$ is 1000, both when the tube is having an open or closed end for a range of dimensionless diffusion time

for a variety of tube geometry and both when the tube is closed and open to the atmosphere in Figs. 5 and 6 when assuming the leakage of these pure fuels takes place in air at atmospheric pressure and temperature. It is to be noted that these figures can also be used if the surrounding air already contains a small concentration of the leaking fuel (i.e., when $C_{02} > 0.0$). An example of such a case is shown in Fig. 7. It can be seen that in such a case the flammable zone expands on the lean side but shrinks on the rich side when compared to the case of pure air.



(*a*)

(*b*)

Fig. 5 Calculated contours of dimensionless diffusion time versus distance for various constant relative concentrations of a diffusing fuel into air for two different typical values of B for a closed tube. Examples are shown how the growth of the flammable zone can be established typically for carbon monoxide and propane in air.

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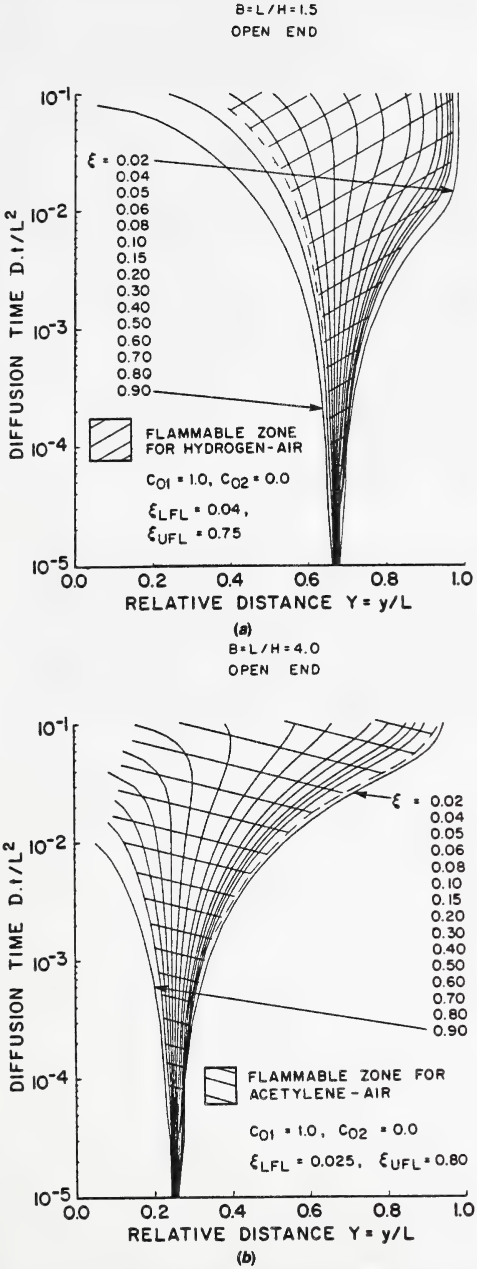

(a)

(b)

Fig. 6 Calculated contours of dimensionless diffusion time versus distance for various constant relative concentrations of a diffusing fuel into air for two different typical values of B for an open-ended tube on the air side. Examples are shown how the growth of the flammable zone can be established typically for hydrogen and acetylene in air.

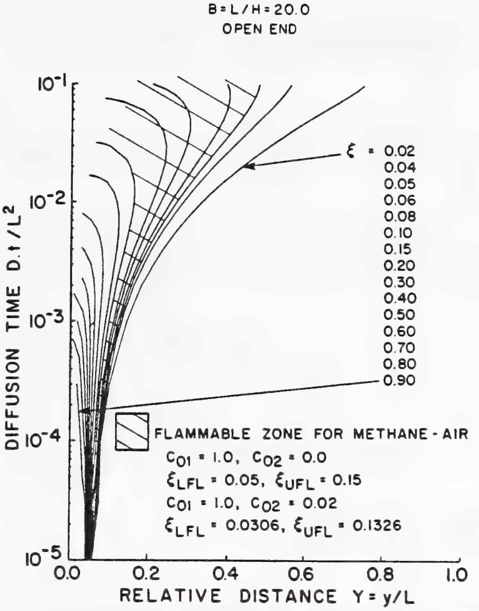

Fig. 7 Calculated contours of dimensionless diffusion time versus distance for various constant relative concentrations of a diffusing fuel into air for a typical value of B for an open-ended tube. Examples are shown how the growth of the flammable zone can be established typically for methane both in air and in an atmosphere containing initially 2 percent methane.

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APPENDIX A

The one-dimensional molecular diffusion process for a non-reactive system assuming constant diffusion coefficient and equi-molar counter diffusion is expressed by

$$\partial c/\partial t = D\, \partial^2 c/\partial y^2 \tag{5}$$

Consider the tube shown in Fig. 1 in which the lower and upper sections are initially filled with homogeneous mixtures of the fuel and air with concentrations C_{01} and C_{02}, respectively. In order to generalize the solution, the following normalized parameters are used:

$\xi = (C - C_{02})/(C_{01} - C_{02})$ = relative concentration
$\gamma = y/L$ = dimensionless distance
$B = L/H$ = dimensionless parameter describing the tube geometry
$\tau = Dt/L^2$ = dimensionless diffusion time

Thus, the diffusion equation and for the two cases of closed and open upper end, the initial and boundary conditions are the following:

$$\partial\xi/\partial\tau = \partial^2\epsilon/\partial Y^2 \tag{6}$$

$$\left.\begin{array}{lllll} & \tau = 0 & 0 < Y < 1/B & \xi = 1 \\ & \tau = 0 & 1/B < Y < 1 & \xi = 0 \\ & \tau > 0 & Y = 0 & \partial\xi/\partial Y = 0 \\ \text{closed end} & \tau > 0 & Y = 1 & \partial\xi/\xi Y = 0 \\ \text{open end} & \tau > 0 & Y = 1 & \xi = 0 \end{array}\right] \tag{7}$$

The solutions of equation (6) are expressed in the following two possible series (17 and 16):

For a closed end:

$$\xi(\tau, Y) = \frac{1}{B} + \frac{2}{\pi}\sum_{n=1}^{\infty}\left[\frac{1}{n}\sin\frac{n\pi}{B}\cos n\pi Y \bullet \operatorname{Exp}(-n^2\pi^2\tau)\right] \tag{8}$$

and

$$\xi(\tau, Y) = \frac{1}{2}\sum_{n=-\infty}^{\infty}\left[\operatorname{erf}\left(\frac{1/B + 2n - Y}{2\sqrt{\tau}}\right) + \operatorname{erf}\left(\frac{1/B - 2n + Y}{2\sqrt{\tau}}\right)\right] \tag{9}$$

For an open end:

$$\xi(\tau, Y) = \frac{2}{\pi}\sum_{n=1}^{\infty}\left[\frac{1}{(n-1/2)}\sin\frac{(n-1/2)\pi}{B}\right.$$
$$\left.\cos(n-1/2)\pi Y \bullet \operatorname{Exp}[-(n-1/2)^2\pi^2\tau)]\right. \tag{10}$$

and

$$\xi(\tau, Y) = \frac{1}{2}\left[\operatorname{erf}\left(\frac{1/B + Y}{2\sqrt{\tau}}\right) + \operatorname{erf}\left(\frac{1/B - Y}{2\sqrt{t}}\right)\right]$$
$$+ \frac{1}{2}\sum_{n=1}^{\infty}(-1)^n\biggl[\operatorname{erf}\left(\frac{1/B + (2n - Y)}{2\sqrt{\tau}}\right)$$
$$+ \operatorname{erf}\biggl(\frac{1/B - (2n - Y)}{2\sqrt{\tau}} + \operatorname{erf}\left(\frac{1/B + (2n + Y)}{2\sqrt{\tau}}\right)\biggr)$$
$$+ \operatorname{erf}\left(\frac{1/B - (2n + Y)}{2\sqrt{\tau}}\right)\biggr] \tag{11}$$

The exponential solutions (8) and (10) require a small number of terms for convergence of the series when having long diffusion times. On the other hand, the error function-based solutions (9) and (11) require a small number of terms when having short diffusion time. Accordingly, in the present work, equations (8) and (10) were used for $\tau > 0.05$ and equations (9) and (11) were used for $\tau < 0.05$. It is also noticed that equation (11) is only valid for an odd number of terms.

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"Investigations of the Generation of Flammable Zones Following the Release of a Fuel Gas into the Atmosphere" (G. A. Karim, M. Kibrya, H. Lam, R. Petela and I. Wierzba); Proceedings of the International Specialists Meeting on Major Hazards in the Transport and Storage of Pressure Liquefied Gases, University of New Brunswick, Fire Science Centre, Fredericton, N.B., August 10-13, 1987.

INVESTIGATIONS OF THE GENERATION OF FLAMMABLE ZONES FOLLOWING THE RELEASE OF A FUEL GAS INTO THE ATMOSPHERE

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ABSTRACT

The results of analytical and experimental investigations of some aspects of the mode of generation of flammable zones following the release of a flammable gas, such as methane or propane, into air are presented. Changes in these zones with the lapse of time are shown for a range of locations away from the fuel source. Moreover, calculated fuel concentration profiles formed following the continuous discharge of a fuel gas from a pressurized container into the open atmosphere are presented. Some guidelines regarding the location and size of the flammable zone in the neighbourhood of some discharges are presented and discussed.

INTRODUCTION

The assessment of fire and explosion hazards arising from the release or seepage of a fuel vapour into an oxidizing atmosphere such as air, whether continuously or intermittently within a reservoir, a pipeline or in the open atmosphere, is a task of paramount importance in the safeguarding against possible damage that may arise as a consequence of such an incident. Clearly, there is a need to be able to establish for any specific configuration and fuel whether a hazardous situation is going to exist, its seriousness and an indication of the extent of its persistence and spread following the release of the fuel.

A criterion that has been widely used in such an assessment is that of the flammability limit which would indicate for any fuel and oxidant the limiting concentration for a flame to propagate within the homogeneous mixture following ignition from a sufficient source of energy. These limits, being the lower and higher limits of flammability, cannot be predicted theoretically and relate only to a homogeneous quiescent mixture under closely specified conditions. The influence of important common operating parameters such as mixture temperature, pressure, movement and turbulence intensity needs to be established experimentally. Very little consistent and sufficiently comprehensive flammability data are available in the literature even for common fuels. Moreover, in practice most hazardous situations involving fuel vapours result usually from the convective release of the fuel into the surrounding environment, resulting in mixtures that are highly nonhomogeneous, nonquiescent and time varying in composition. The treatment of such situations is significantly more complex and time consuming than those involving homogeneous mixtures.

Clearly, the prediction of the mode of development of a fuel-air mixture with respect to time and space is a very favourable candidate for analytical modeling. However, at the present time the development of a suitable comprehensive theoretical model is untenable. Nevertheless, simple situations can be considered which can permit analysis and provide a useful insight into the formation of flammable mixtures albeit for specific situations.

The present contribution considers some aspects relating to fuel-air mixture formation in specific instances involving the discharge of a fuel vapour into air and the subsequent generation of flammable zones both under steady and transient conditions. It is then shown that for these conditions, guidelines can be formulated regarding the location and size of the hazardous zone in the neighbourhood of such fuel discharges.

CHANGES IN THE FUEL CONCENTRATION WITH TIME DUE TO MOLECULAR AND CONVECTIVE DIFFUSION

The concentration of a fuel vapour following the release of a finite quantity of fuel into still air, in the absence of the effects of gravity or pressure and temperature gradients can be predicted well as a function of time and space. The well known Fick's Law Equation for transient molecular laminar diffusion (ref. 1) can be applied then with confidence and appropriate efficient solutions for the equation are available in the open literature (ref. 2). For example, the release of heavy fuel vapours such as propane, chlorine, etc. upwards into air, light fuels such as methane, ammonia, hydrogen, etc. downwards into air or gases in general horizontally into still air, can be established reliably by applying the assumption that molecular diffusional processes are involved.

As an example, the upward or horizontal transient diffusion of the heavy gaseous fuel propane from a cylindrical section of a finite volume 'H' located at one end within a long open pipe of air at atmospheric pressure and temperature is considered and shown schematically in Fig. 1. The changes in the volumetric concentrations of the propane in air with time following the initiation of diffusion are shown in the figure at three typical locations along the open tube. It is evident that this transient diffusional process is very slow and would result in flammable mixtures over a significant region of the tube, away from the initial interface between the fuel and air, existing for a very long period of time. Furthermore, depending on the specific configurations employed, sufficiently rich mixtures can be formed within regions of the tube to produce for some time non-flammable conditions when the fuel concentration is greater than the corresponding rich flammability limit of propane in air (≈ 9.0%). Similar type of calculations can be made for situations involving the diffusion of propane into air within a closed pipe of finite length. As shown in Fig. 2, relating to a typical case of the release of a finite quantity of propane of a cylindrical volume 'H' shown schematically, upwards into air at constant temperature and pressure under molecular diffusion conditions, the diffusion time needed to effect homogenous and thorough mixing of the fuel and air is very long indeed. Moreover, a changing concentration gradient will exist along the tube over much of the time prior to achieving near equilibrium concentrations. At locations near the region of the interface between the fuel and air, there can exist a fuel concentration build-up over a significant period of time to extents well beyond the ultimate fuel concentration at equilibrium. Accordingly, although the release of the fuel in such a tubular reservoir may eventually produce fuel concentrations in air outside the flammability limits, care needs to be taken as there will exist for a significant period of time flammable zones in part of the reservoir.

Molecular diffusion, which tends to be extremely slow, is not commonly encountered in most practical situations involving the dispersal and mixing of a fuel vapour into air. The application of the molecular diffusional approach to the spread of a fuel vapour into air, when the conditions required for achieving such a diffusion do not fully exist, can lead to gross errors in estimating the development of the fuel concentration in air. For example, such conditions can be under the influence of bouyancy effects, whether due to the release of a light gas upwards into air or due to the presence of temperature non-uniformities within the medium. Experiments reported by Karim and Lam (ref. 3), involving the transient convective diffusion of methane into air at constant pressure and temperature within a long vertical tube, established the concentration changes of the methane with respect to time and space using non-intrusive ultrasonic gas analysis. As shown in Fig. 3, the employment of the transient molecular diffusional approach produces values for the fuel concentration that are drastically different from those established by direct measurement. The actual convective process as measured, is some orders of magnitude faster and can produce peak concentrations that are higher than those predicted by the simple theory. Moreover, accounting for bouyancy effects but retaining the laminar transport processes assumption can still lead to unacceptably large deviations from the corresponding experimental values (ref. 4).

It is evident that the mixing process due to the convective diffusion of the light fuel is speeded up due to the onset of turbulence and instabilities, particularly at regions near the initial fuel-air interface and at the early

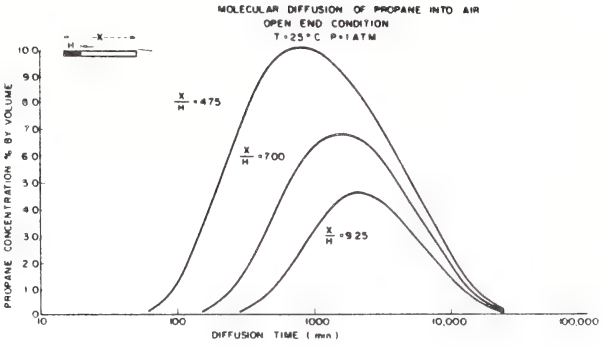

Fig. 1. Calculated volumetric concentration of propane as a result of one-dimensional molecular diffusion into air within an open cylindrical tube versus time at atmospheric temperature and pressure for three locations (X) along the tube. Molecular diffusion is initiated promptly at time zero from a cylindrical segment (H) at one end containing propane, as shown schematically.

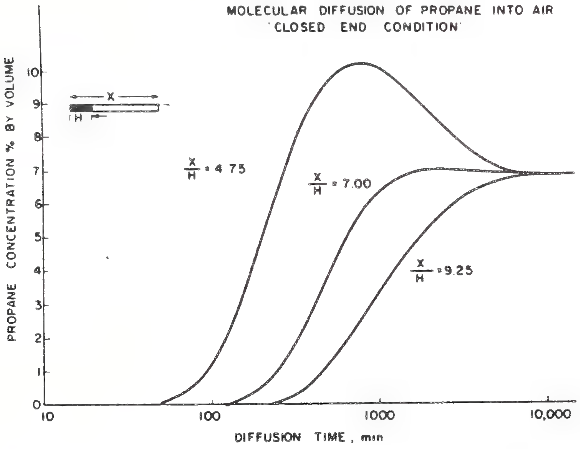

Fig. 2. Calculated volumetric concentration of propane as a result of one-dimensional molecular diffusion into air within a closed cylindrical tube versus time at atmospheric temperature and pressure for three locations (X) along the tube. Molecular diffusion is initiated promptly at time zero from a cylindrical segment (H) at one end containing propane, as shown schematically.

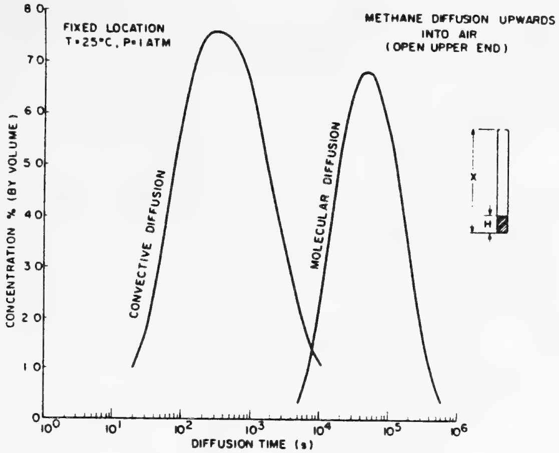

Fig. 3. A typical variation with time of the measured concentrations of methane diffusing upwards into air within a long open cylindrical tube at atmospheric temperature and pressure at a specific location along the tube ($\frac{X}{H}$ = 7.0). The corresponding calculated concentration of methane on the basis of molecular diffusion is also shown. Diffusion is initiated promptly at time zero from a cylindrical segment (H) containing methane, at one end, as shown schematically.

stages of the mixing process (ref. 5). Hence, if a simple model for diffusion is to be retained, an effective eddy diffusivity needs to be used that is orders of magnitude larger than the value of the molecular diffusivity. Analysis of the experimental data of Karim et al (ref. 6) for the diffusion of methane into air within a long tube showed that for good agreement of predicted methane concentrations with the corresponding experimental values, such an effective eddy diffusivity needs to be varied during the course of the diffusion. Its value needs to be reduced somewhat with the progress of diffusion and time and at regions away from the initial interface. Furthermore, it can be shown (ref. 7) that, immediately after the initiation of the diffusion, the presence of radial diffusion cannot be ignored, resulting in the need for at least a two-dimensional axisymmetric model for the diffusion, instead of the simple one-dimensional approach employed. Figure 4 shows the extent of agreement of the predicted times for the arrivel and subsidence of the flammability limit concentration of methane in air at various locations along the long vertical cylindrical pipe with the corresponding measured values. Good agreement of the predicted values with experiment is evident when appropriate different effective diffusivity values are employed.

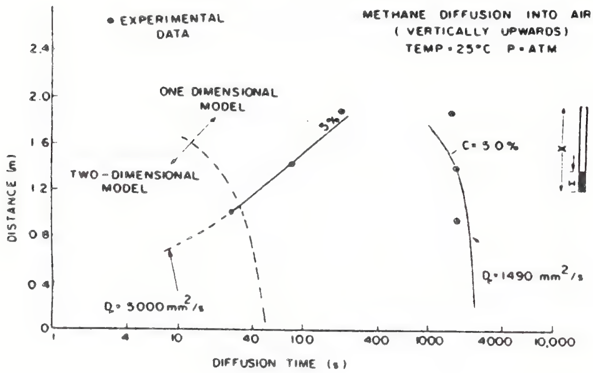

Fig. 4. Typical calculated times required to achieve at various locations along a vertical cylindrical tube a concentration of 5% by volume of methane in air following the release of the methane upwards within the tube and when employing two different effective diffusivity values for the early and later parts of the one-dimensional diffusion. Methane is initiated promptly at time zero from a cylindrical segment at one end. Experimental data are shown together with the region where a two-dimensional diffusion model is needed for good agreement with experiment.

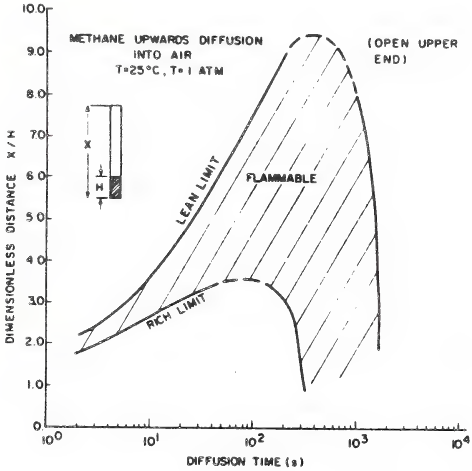

Fig. 5. Changes in the boundaries of the flammable zone with time following the convective diffusion of methane into air within a circular vertical tube. The initial location of the interface between the fuel and air is 0.24m.

Continuing work suggests that the value of such a variable effective eddy diffusivity at any location is a function of the local transient concentration gradient. The employment of such an approach, as shown in Fig. 5, can thus yield realistic information as to the formation, growth and decay of flammable zones within the vertical tube being considered and shown schematically.

THE GENERATION OF FLAMMABLE REGIONS AHEAD OF A FUEL JET ISSUING INTO AIR

The rapid discharge of a fuel from a container into a still atmosphere through an opening or an orifice due to a pressure difference will generate a flammable region ahead of the issuing turbulent jet. Examinations of the extent of this region can be made analytically in simple cases for a number of fuels and configurations of the surrounding environment.

The concentration of a fuel at any point within a steady fuel jet issuing into air depends on the entrainment rate of air by the issuing fuel jet. The axial and radial concentration profiles of the fuel can be calculated by employing well established jet theories (ref. 8). Hence, the locations of the lower flammability, stoichiometric or the higher flammability limit zones can be established. Moreover, on ignition of the jet, the resulting flame is associated with certain concentrations of the fuel in air, normally that of the stoichiometric value. Accordingly, the possible length and location of the flame relative to the point of emission can be established analytically.

If the surrounding air were to contain some fuel vapour in small concentrations below the corresponding flammability limit, such as due to a gradual build-up of fuel vapour in the environment of the immediate vicinity of the fuel jet, then the jet will entrain this additional fuel with the air. Accordingly, the concentration of the total fuel at every point within the region of the jet and its associated stoichiometry will change as the concentration of this additional fuel is altered. Consequently, the shape and size of the flame and the flammability limit zones will change.

From the concentration profiles of air and fuel, the equivalence ratio, which is the local fuel to air ratio relative to the corresponding stoichiometric value, can be calculated. Hence, moving across the jet region, the concentration corresponding to the stoichiometric, lower or higher flammability limits are located. Therefore, although the general shape of the fuel concentration distribution will remain essentially the same, the concentration of the total fuel at any point will be increased somewhat due to the induction of the surrounding fuel with the air. The local concentration of the air will be reduced by the same amount. Consequently, the fuel to air ratio at that point will become correspondingly higher. The stoichiometric envelope would also move outwardly, resulting in increases in the flame width and length.

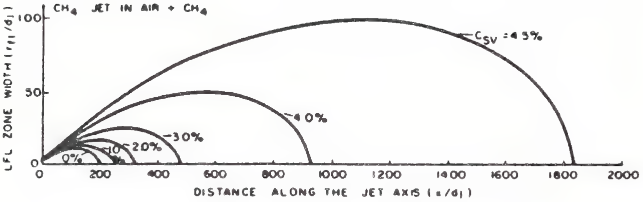

Fig. 6. The shape and size of the lower flammability limit (L.F.L.) envelope formed around a methane turbulent jet issuing in air or air containing various concentrations of methane as % by volume of the surroundings.

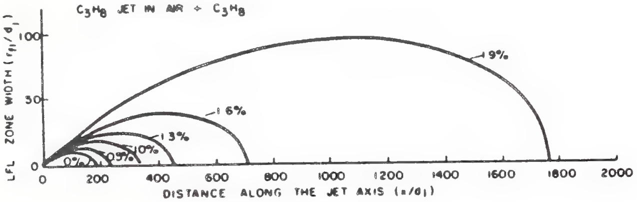

Fig. 7. The shape and size of the lower flammability limit (L.F.L.) envelope formed around a propane turbulent jet issuing in air or air containing various concentrations of propane as % by volume of the surroundings.

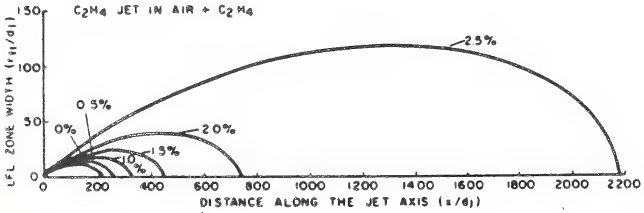

Fig. 8. The shape and size of the lower flammability limit (L.F.L.) envelope formed around an ethylene turbulent jet issuing in air or air containing various concentrations of ethylene as % by volume of the surroundings.

Figure 6 shows the boundaries of the lower flammability limit zone following the mixing of a fuel, methane, into air from a fuel jet issuing into unrestricted environment of air. The extension of these boundaries due to the presence of small concentrations of methane premixed in the surrounding air is also shown. It can be seen that the flammable zone extends both in length and width very markedly, as the background fuel concentration increases. Of course, a continued increase in the fuel concentration in the surroundings will lead eventually to rendering the whole surroundings flammable. Figures 7 and 8 show similar plots for propane and ethylene jets respectively, discharging into air or air containing some premixed fuel. Accordingly, a gradual build-up of the fuel concentration in the environment in which the fuel jet is discharging, will render such a discharge more hazardous and the consequences of uncontrolled ignition of the fuel jet more severe (ref. 9).

FLAMMABILITY AND FLAME PROPAGATION CHARACTERISTICS UNDER STRATIFIED MIXTURE CONDITIONS

The apparent limiting mixtures established under convective diffusion conditions involve neither a homogeneous mixture nor a quiescent state, since vigorous convection may take place. This convective diffusion, particularly just after the sudden commencement of the diffusional process, can involve small scale turbulence influencing the local state and concentration of the fuel at the ignition location. Hence, a lower concentration of fuel than that of the established flammability limit, is needed to permit combustion (ref. 10).

When determining the flammability limit for a homogenous quiescent mixture, the resulting flame is required to propagate the whole length of the test tube to consider the mixture flammable (ref. 10). However, in stratified mixtures, the resulting flame propagates shorter distances merely to the location of the limit mixture. Hence, a relatively shorter flammable zone may be generated around the ignition source location with a concentration lower than the lean limit for homogeneous mixtures. In the presence of a positive concentration gradient, the flame front moves to regions of higher fuel concentrations releasing enough energy to sustain and even accelerate the flame in the case of lean mixtures.

The variation of the apparent lean limit for the methane-air stratified mixture within a vertical tube with the local concentration gradient is shown in Fig. 9. As would be expected, the observed values are the same whether the tube end was open or closed (ref. 3).

Locations closer to the initial interface between the fuel and air have higher concentration gradients. Thus, a lower local concentration is needed to initiate a flame at this location than away from it. Similarly, the corresponding values of the concentration of the fuel in air at locations where the flame appears to die out because of encountering a too rich mixture, are somewhat higher than the corresponding value of the rich flammability limit when established for homogeneous quiescent mixtures and downward flame propagation. The induced small scale turbulence due to the local concentration gradient probably contribute to this slightly higher rich limit.

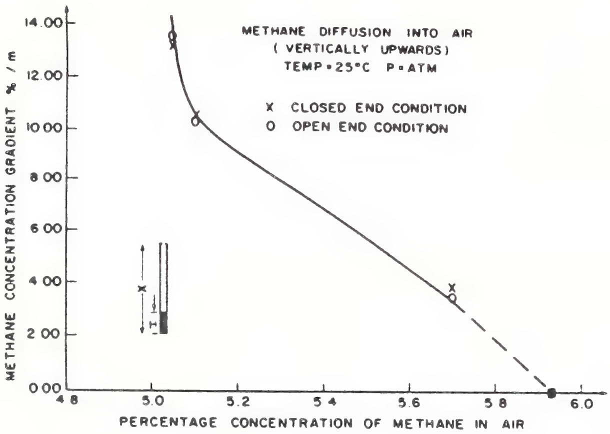

Fig. 9. Experimentally established lean flammability limit of stratified methane in air mixtures within a vertical circular pipe. Stratification is formed following the convective diffusion of methane upwards into air (ref. 3).

The apparent flame propagation rate within the stratified methane-air mixtures were examined. At the minimum diffusion time for generating an 'ignitable' mixture at any specified location, a flame was initiated with sparking periodically the mixture at that location. The resulting apparent flame speed values are shown in Fig. 10 as a function of the corresponding local methane concentration. It can be seen that results involving stratified mixtures have the same general trend as those for homogeneous mixtures. However, the apparent flame speeds for both accelerating and decelerating flames within the stratified mixtures tended to be higher than those encountered under homogeneous conditions for the same methane concentration (ref. 5).

The dominant factor for the enhanced flame speed observed within the rich region is the corresponding change in the amount of heat generated at the flame front and the temperature of the burned gases. For homogeneous mixtures, the amount of energy released at the flame front and the flame temperature remain essentially constant. A possible reason for the observed difference in the apparent flame speeds within the lean region, is the convective effect of the increasing methane concentration in the direction of flame propagation. Hence, the flame front will experience a higher concentration than expected and prop-

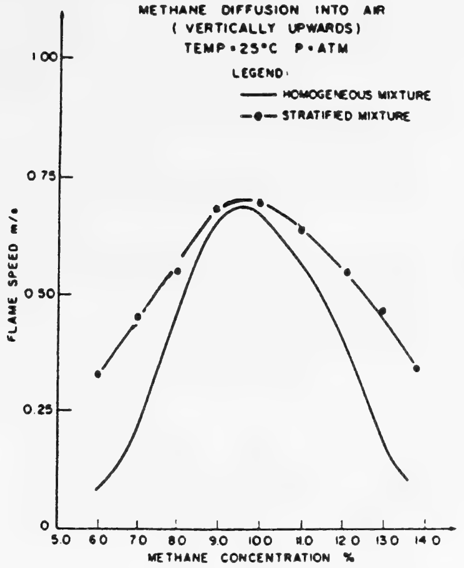

Fig. 10. The measured apparent flame speed propagating downwards within a stratified mixture of methane and air formed by the upward convective diffusion of methane within a vertical tube as a function of the local methane concentration. The corresponding flame speed for homogeneous mixtures, in the same apparatus, is also shown (ref. 3).

agate at a higher speed. However, for propagating flames with decreasing velocity from the stoichiometric mixture to the rich limit, the rate of heat transfer to the unburnt mixture would be increased through relatively greater conduction and radiation heat transfer due to the higher temperature difference. The flame temperature would be higher for stoichiometric than for rich mixtures and the enhanced preheating of the unburnt gases, compared to the homogeneous conditions, would result in faster local flame speeds compared to those under homogeneous conditions.

ACKNOWLEDGEMENT

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Experimental and Analytical Investigation of the Convective Diffusion of Methane Into Air

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The concentration changes with time and space of methane gas were examined following its release into air at atmospheric pressure within a long smooth cylindrical tube of 2.5 in. (63.5 mm) diameter, taking into account the combined effects of molecular diffusion and natural convection. The concentration profiles as a function of diffusion time at specified regions of the tube was determined by measuring the velocity of sound in the medium along a tube diameter using an ultrasonic gas analyzer. Comparative tests involving carbon dioxide, neon and helium were also made. It was shown that the experimental concentration profile for the diffusion of methane observed may be reproduced using the simple one-dimensional classical diffusion equation when the molecular diffusivity was replaced by a very much larger effective eddy diffusivity that appeared to depend on the diffusion time and local concentration gradient.

Introduction

Combustion within a stratified atmosphere of air and methane is frequently encountered in practice. Examples can be found inside combustion chambers, furnaces and internal combustion engines, around leaking fuel storage tanks or pipelines and within gas pockets in coal mine galleries. The development of a flame following ignition within such atmospheres would depend on the stratification patterns of the gaseous fuel in the air. Such stratification is formed mostly by a diffusion phenomenon where molecular diffusion is often combined with turbulence and natural convection effects. The formation of such stratified atmospheres is not adequately known and most modeling attempts, though involving complicated numerical methods, could claim only approximate solutions to the problem (Jensen and Finlayson, 1980).

Early workers (Coward and Georgeson, 1941) investigated theoretically and experimentally the diffusion of methane in air employing a coefficient of interdiffusion that is a function of both temperature and pressure. Later, other factors such as the air speed, pressure release (Titman, 1959), buoyancy effects (Bakkle, 1959; Bakkle and Leach, 1962; Leach and Barbero, 1961), and ventilation rates (James and Purdy, 1962) were examined and the effects of the density gradient in a system inherent in natural convection was discussed (Turner, 1973; Velarde and Normand, 1980; Berge, 1982).

The characteristics of flame propagation within stratified atmospheres of methane in air were investigated experimentally (Meerbach, 1963; Liebman et al., 1969; Liebman et al., 1971; Perlee at al., 1964; Karim and Tsang, 1974; Badr and Karim, 1984) and theoretically (Chu, 1952; Schalla, 1961). A model for convective diffusion was also developed (Tsang, 1974) and its theoretical solution was compared with the experimental concentration measurements deduced using an ultrasonic gas analyzer (Karim et al., 1976). This model after modification was used to investigate the behavior of flame propagating within the stratified combustible mixture (Badr, 1977; Panlilio, 1981; Karim and Lam, 1986).

The present paper considers the case of upward diffusion of a buoyant gas such as methane into air within a vertical smooth long circular tube. The concentrations of the diffusing gas with respect to time were measured at different locations and then used to evaluate effective values for the apparent diffusion coefficient, eddy diffusivity, under such stratified mixture conditions.

Experimental Apparatus

A long circular smooth flame tube 2.5 in. (63.5 mm) i.d. consisting of a 9-ft (2.75-m) upper air section and an 8-in. (0.203-m) long fuel section was employed. The large length-to-diameter ratio was used so as to approximate to a one-dimensional situation. The tube assembly and associated apparatus are shown schematically in Fig. 1.

A quick action thin smooth flat plate full area shutter was connected between the two sections of the tube containing the air and methane. Upon rapid opening of the shutter, natural convection and molecular diffusion allowed both gases to interdiffuse within each other, generating in time a stratified atmosphere. The concentrations at any point within the flame tube changed with time. An electric spark of adequate and constant energy was employed to initiate after a certain specified diffusion time, flame propagation, when needed.

An approach involving the use of fiber optical cables was developed to determine the apparent speed of the propagating flame within the tube (Karim and Badr, 1976).

To determine experimentally the instantaneous average gas

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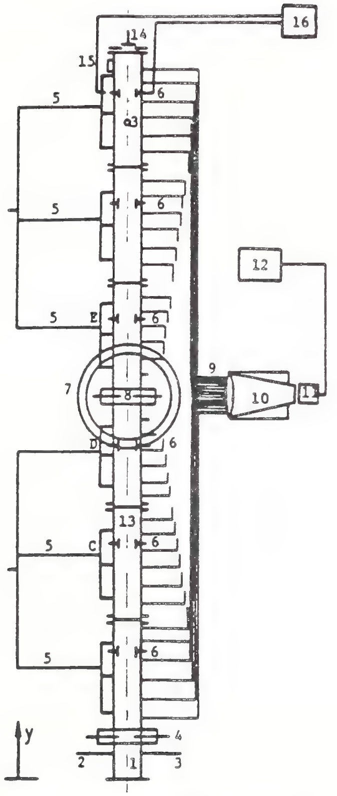

Fig. 1 Schematic diagram of the apparatus employed

1 Fuel section (lower compartment)
2 Fuel inlet
3 Evacuation line
4 Plate valve/shutter
5 Air inlet manifolds
6 Transducer port/movable ignition source
7 Rotating (inclination) disk
8 Supporting plate
9 Fiber optics
10 Optical system for light concentration
11 Photo diode
12 Photo diode output recorder
13 Air section (upper compartment)
14 Nozzle valve
15 Rupture diaphragm relief valve
16 Ignition circuit

concentration along a tube diameter at different test locations, a "sonic gas analyzer" of our design was employed. The velocity of sound in a gas mixture is a function of its density. Therefore, through the passage of an ultrasonic pulse across a tube diameter at the test location, it is possible to determine the concentration of the species in a binary homogeneous mixture of gases from the measurement of velocity of sound in the mixture under isothermal conditions. Changes in the concentrations due to diffusion with time can thus be established. A calibration curve was obtained by measuring the velocity of sound for accurately known and prepared, homogeneous methane-air mixtures within the flame tube under constant ambient pressure and temperature. This experimentally derived curve was used subsequently for determining the changes

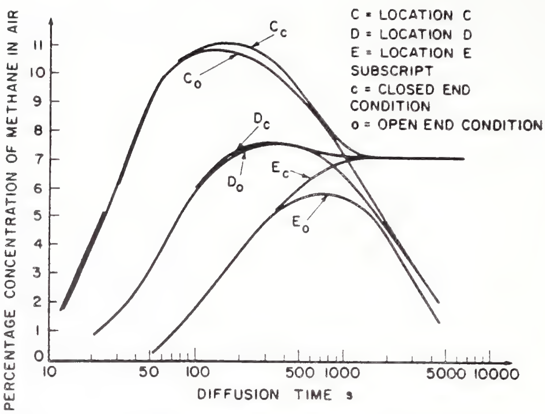

Fig. 2 Measured concentration of methane by volume at three locations along the tube C, D and E, both when open and closed, located at distances of 30.0 in. (0.762 m), 48.0 in. (1.220 m) and 66.0 in. (1.676 m), respectively, away from the position of the plate valve separating initially the two gases at atmospheric pressure and temperature. The fuel section was 8 in. (203 mm) long.

in the concentration with time within the tube. The mean time interval for the ultrasonic pulse (40 kHz) to travel across the tube was obtained by recording 50 readings using a data acquisition system. The associated error was assessed to be ±0.08 percent.

The conditions considered in this work involved the stratification of methane in air through the rapid opening of a full-area, thin and smooth plate valve. The stratification of methane in air in the radial direction, although it may exist, was assumed to be initially small compared to the axial gradients away from the initial inter-phase region. It is acknowledged that if the methane was released from a point or a line source into air simulating a leak, then this view may need to be modified. Similar experiments involving the release of the more buoyant helium into air indicated that such a one-dimensional assumption may be unjustified. No other similar tests were carried out in apparatus having a substantially different size or geometry from the one employed in this investigation.

Changes in the Concentration With Time Due to Convective Diffusion

The concentration of methane in air as a function of diffusion time at a number of locations along the flame tube was established using the ultrasonic gas analyzer. Figure 2 shows typical experimental results for the conditions, both when the tube was closed and when it had an open upper end, at three locations, C, D and E, at distances of 30.0 in. (0.762 m), 48.0 in. (1.220 m) and 66.0 in. (1.676 m), respectively, away from the plate valve position separating the two sections. Although each individual curve displayed in general a similar trend to that expected from the solution of the one-dimensional molecular diffusion problem with an open end condition, the actual diffusion time observed is drastically faster.

Nomenclature

C, D, E = locations along tube where observations made
c_1 = concentration within lower section (mol/mol)
c_2 = concentration within upper section (mol/mol)
D_c = effective diffusion coefficient (m^2/s)
erf = error function
h = length of fuel section (m)
L = whole length of tube (m)
n = number of terms
t = time (s)
x = distance along tube (m)

Subscripts

o = initial conditions

Keeping initially the ultrasonic transducers at a fixed location (1.220 m away from the separating valve, i.e., 1.420 m from the bottom end of the tube), the diffusion of different gases in air was investigated. The aim was to obtain the concentration profiles and examine the effect of having different gases with different molecular weights on the resulting concentration profiles when the upper end of the tube was open to the outside atmosphere. The test gases were chosen in such a way so as to allow the effects of molecular diffusion and natural convection due to different extent of buoyancy, to be studied. Plots of the instantaneous concentration profile against diffusion time are shown in Figs. 3 and 4 for four binary mixing systems of carbon dioxide, methane, neon and helium when diffusing upwards individually into air. The diffusion time was defined to be equal to zero, when the shutter separating the gas container from that of air was released for the initiation of the diffusive mixing process.

The gas concentration increased initially at a fast rate because of the severe concentration gradient encountered, then reached a peak concentration. Later on, it continued to decrease very gradually due to the diffusion and dissipation of the gas up the long tube and through the open end into the atmosphere. In the CO_2-air system, mixing took place purely by molecular diffusion, which is relatively very slow. For the other gases considered, both molecular diffusion and natural convection were involved in the mixing process which could have then also involved small-scale turbulence that accelerated the transport and mixing processes.

In systems involving the diffusion of methane, helium or neon into air, buoyancy is the dominant driving force speeding up the mixing process very significantly relative to that under the influence of ordinary molecular diffusion. The trend of the resulting concentration profiles with diffusion time for these cases tends nevertheless to remain essentially similar.

Following the opening of the separating shutter, the test gas initially would diffuse very rapidly into the air since the density difference between the released gas and the air is very large. The peak concentration is reached sooner with the lesser dense gas relative to air except, as shown in Fig. 4, in the case of He. This trend is believed not to be due to the inadequacy of the ultrasonic gas analyzer concentration measurement, but is due to the nonhomogeneity of the diffusing binary mixture at the test cross section and the breakdown of the assumption of a one-dimensional diffusion. The fast diffusing helium tends to stratify both laterally as well as radially. A simple calculation would show then that the time required for the sound signal to travel along a tube diameter through such a stratified mixture would be longer than the corresponding time for the equivalent homogeneously mixed case at the same cross-section. Thus, it would indicate misleadingly a lower concentration for the helium than for homogeneous diffusion. This observation with the highly buoyant and diffusive helium into air emphasizes the need to restrict the one-dimensional treatment to moderately buoyant gases or to stations well away from the separation between the gas and air or to later times

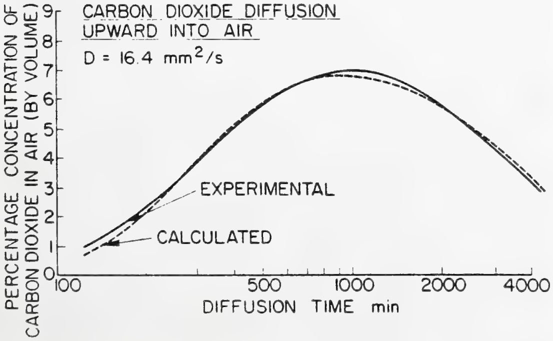

Fig. 3 Concentration profile versus diffusion time obtained at location D, 56 in. (1.422 m) away from the bottom of the tube for the binary system of carbon dioxide diffusing upwards into air—open upper end condition and carbon dioxide section of 8 in. (203 mm) long. The calculated profile is also shown.

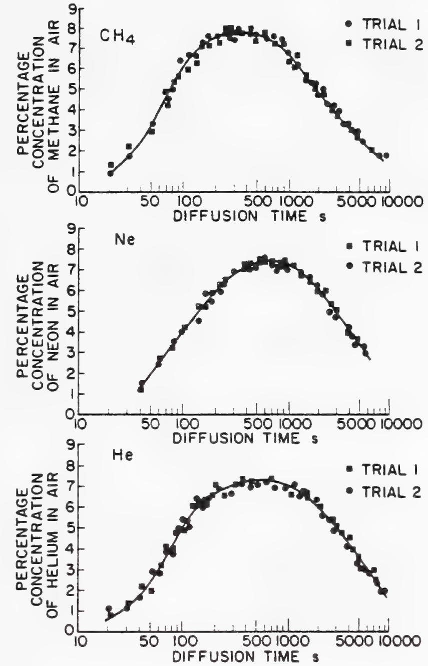

Fig. 4 Measured concentration profile versus diffusion time obtained at location D, 48 in. (1.220 m) away from the position of the plate valve separating initially the gas and air for three different cases involving the diffusion of methane, neon and helium into air at atmospheric pressure and temperature

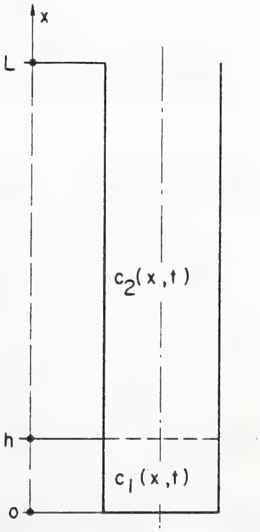

Fig. 5 Schematic of the two sections of the tube

when the density differences have subsided. Otherwise, the analysis will be both complex and multi-dimensional.

As the methane was released into the air by opening the plate shutter, the concentration gradient and hence the density gradient, at the interface of the methane and air, was extremely high initially. This inevitably led to turbulent mixing and associated eddies tending to dissipate such large gradients. For locations further away from the shutter, the scale of turbulence would diminish and the size of eddies would become smaller, smoothing out and reducing such a sharp concentration gradient at the shutter. The scale of turbulent mixing and the size of the associated eddies in turn depend mainly on the concentration gradient. Furthermore, the variation in the concentration gradient, as well as the scale of turbulent mixing, can explain the trends observed in Fig. 2, where the location nearer to the shutter had a larger slope at the early diffusion time and reached the peak concentration earlier than locations further away.

As seen from Fig. 2, for the closed end condition, after a certain diffusion time, the concentrations at any fixed location differed from those obtained for the open end situation. The value increases expectedly as the distance between the test location and the closed upper end increased. The furthest location tested needed the longest diffusion time to sense the boundary difference. The final concentration at all locations leveled off to an asymptotic value corresponding to that when methane was mixed homogeneously with the air within the entire tube.

Analysis of Experimental Results

To compare the measured concentrations with those that may be obained by a theoretical model, let the diffusion process be considered first simply for a constant effective diffusion coefficient D_e. After the elapse of a time period t, one-dimensional diffusion occurs both in the bottom and upper parts of the vertical tube (Fig. 5). To calculate the gas concentration c_1 in the lower part, consider the differential equation for $0 \leq x < h$

$$\frac{\partial c_1}{\partial t} = D_e \frac{\partial^2 c_1}{\partial x^2}$$

with boundary conditions

$$\frac{\partial c_1}{\partial x} = 0 \quad \text{for } x=0 \quad \text{and} \quad t \geq 0$$

and $$c_1 = c_o \quad \text{for } 0 \leq x < h \quad \text{and} \quad t=0$$

Similarly, for the gas concentration c_2 in the upper part of the tube, $h \leq x \leq L$, the boundary conditions are

$$c_2 = 0 \quad \text{for } x \geq h \quad \text{and} \quad t=0$$

and

$$c_2 = 0 \quad \text{for } x=L \quad \text{and} \quad t \geq 0$$

Additionally, for the interface between the bottom and upper parts, the following condition applies:

$$c_1 = c_2 \quad \text{for } x=h \quad \text{and} \quad t>0$$

and

$$\frac{\partial c_1}{\partial x} = \frac{\partial c_2}{\partial x} \quad \text{for } x=h \text{ and } t>0$$

Applying the Laplace transform method, the following solutions are obtained:

$$c_1 = c_o - \frac{c_o}{2} \sum_{n=0}^{\infty} (-1)^n \left\{ 4 - \operatorname{erf}\left[\frac{2Ln+h-x}{2\sqrt{D_e t}}\right] - \operatorname{erf}\left[\frac{2Ln+h+x}{2\sqrt{D_e t}}\right] - \operatorname{erf}\left[\frac{2L(n+1)-h-x}{2\sqrt{D_e t}}\right] - \operatorname{erf}\left[\frac{2L(n+1)-h+x}{2\sqrt{D_e t}}\right] \right\} \tag{2}$$

and

$$c_o = \frac{c_o}{2} \sum_{n=0}^{\infty} (-1)^n \left\{ -\operatorname{erf}\left[\frac{2Ln-h+x}{2\sqrt{D_e t}}\right] + \operatorname{erf}\left[\frac{2Ln+h+x}{2\sqrt{D_e t}}\right] + \operatorname{erf}\left[\frac{2L(n+1)-h-x}{2\sqrt{D_e t}}\right] - \operatorname{erf}\left[\frac{2L(n+1)+h-x}{2\sqrt{D_e t}}\right] \right\} \tag{3}$$

Both equations (2) and (3) are applicable in the whole range $0 \leq x \leq L$. However, better convergency is for values of c_2 for which $n > 4$ is sufficient.

In the simple case of the upward diffusion of CO_2 into air, employing a diffusion coefficient D = 16.4 mm^2/s, being the molecular diffusion coefficient of CO_2, good agreement between the experimental and calculated results, as shown in Fig. 3, was displayed. This, apart from reflecting well about the reliability of the ultrasonic gas analyzer, confirms clearly that the upward diffusion of CO_2 in air is purely molecular, without any convective effects.

The simple approach of equations (1) and (2), for convenience, may be suggested to apply to cases involving natural convection when the effective diffusion coefficient D can be recognized then as being the "eddy diffusivity." Comparison of the measured concentration of methane at any location with the concentration calculated on this basis showed that the eddy diffusivity values needed were three to four orders of magnitude larger than the corresponding molecular diffusion values. It was further found that the calculated concentration profile when using a single constant eddy diffusivity value could not fit the entire experimental curve. A variable eddy diffusivity was needed which was a function of diffusion time and hence the local concentration and its gradient. As shown typically in Fig. 6 for relatively small diffusion times, the curve with a high eddy diffusivity (0.0050 m^2/s) fitted the experimental curve well. As the diffusion time increased, a lower eddy diffusivity (0.0012 m^2/s) appeared to provide a better fit. Towards the end of the diffusion process, a further decrease in the value of the eddy diffusivity was needed to match the experimental curve.

The large value for the eddy diffusivity, particularly at the early part of the process, is to be expected since the eddy diffusivity depends on the mixing length, which in turn depends

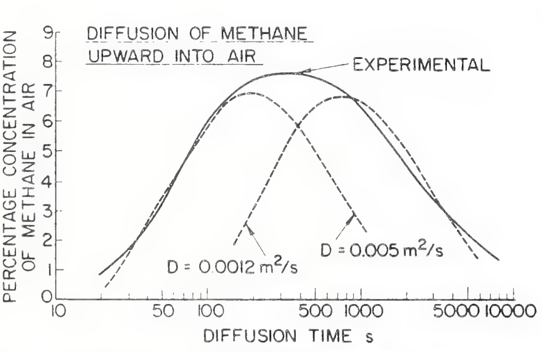

Fig. 6 Comparison of the experimental concentration profile with the corresponding predicted profiles using two different constant values of eddy diffusivity at location D for the binary system of methane diffusing upwards into air within an open tube

on the intensity of turbulence created by the natural convection process. Initially, the intensity of turbulence would be higher as a result of the larger density gradient existing than generating a bigger buoyant force and larger eddies. Later on, as the density gradient became smaller, the turbulence intensity became smaller and so did the eddy diffusivity. Hence, for long diffusion times, curves with smaller eddy diffusivity values would fit experimental results better. The value of the eddy diffusivity tends to behave in the same manner as the average concentration gradient with diffusion time. A high average concentration gradient would thus correspond to a high value of eddy diffusivity.

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SOME GENERAL GUIDELINES FOR THE CONTROL OF FIRE AND EXPLOSION WITHIN INDUSTRIAL ESTABLISHMENTS

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SOME NOTES WRITTEN FOR
GOVERNMENT OF ALBERTA OCCUPATIONAL HEALTH
AND SAFETY HERITAGE GRANT PROGRAM

AGREEMENT #85-51-R
October, 1989

Some of the main steps that need to be taken sequentially to guard against the risk of fire and explosion in industrial fuel installations are listed and discussed. Moreover, the steps to be followed for combating and fighting a fire once it develops are also set up.

Introduction

Numerous national and international standards have been formulated over the years to aid in the planning of prevention measures for fires and explosions in various industrial installations and situations (e.g. 1) There is, of course, in addition, a great wealth of relevant technical and scientific information that already exists in the open literature that needs to be consulted, understood and applied in the design and operation of various equipment and processes.

There is a need for exercising much care and precision in using relevant terms relating to fire and combustion. For example, the terms flammability, inflammability, explosion or explosible limits tend to be used interchangeably. However, it is preferable to use the term flammability limits only for gases and vapours and when relating to homogeneous mixtures in air under specified conditions. For dust, powders, or fuel mist of droplets, the term explosion limits is normally applied. Of course, the term is used to describe vapour, a substance that can become liquid at standard conditions (0^oC, 100 kPa) while a gas remains in the gaseous state at ambient conditions. Explosion is the phenomenon resulting from the very rapid release and expansion of gases. It can occur from events that include mechanical, physical or chemical changes, such as due to the failure of a pressure vessel or the rapid energy release due to the exothermic reaction of chemical explosives. Higher pressures developed as a result of explosion are primarily due to the increase in temperature. Accordingly, peak explosion pressures tend to occur around the stoichiometric mixture region. Deflagration is a term used to describe the propagation of a reaction front within a flammable mixture following ignition at subsonic velocities, such as with flame propagation. However, detonation is a very rapid exothermic reaction that proceeds within the mixture at supersonic velocities accompanied by an intense shock wave. The flash point of a liquid fuel refers to the minimum temperature at which a sufficient amount of fuel vapour is given to form an ignitable mixture with air in a specified vessel used in the standard test (2). The fire point, which is slightly higher than the flash point, corresponds to the temperature at which a liquid continues to burn once it is ignited under standard test conditions. The numerical values of these limits and points are to be used merely to serve for comparative purposes of various fuels in relation to continued fire spread following ignition. In practical settings which almost always involve different conditions from those of the standard tests, different effective flash point values may be involved. The standard flash point according to the closed cup method (2) increases linearly with the initial boiling point of the fuel (3). Sprays, mists and foams of liquid fuels in air which involve widely varying fuel-air mixture conditions within the space being considered can be flammable below the flash point, as shown schematically in Figure 1 (4). A decrease in pressure can lower the flash point of a liquid fuel significantly due to the greater ease with which the fuel vapour is generated and mixed with the air.

The autoignition, self ignition or spontaneous ignition temperature is the minimum temperature at which ignition takes place within a combustible mixture. The value even for any specific fuel will vary depending on many factors influencing the physical and chemical processes during preignition and the ignition delay, e.g. concentrations, temperature, pressure, the presence of catalytic effects, fluid motion, turbulence, the time required for producing successful ignition, etc. Table I shows a typical listing of the main combustion characteristics of some common pure gaseous and liquid fuels (5). The values of the corresponding properties of common fuels that are made up of mixtures of a wide range of fuels such as gasoline, diesel fuel, kerosine, natural gas or L.P.G. would vary widely depending on the composition.

Protection of Fuel Installations Against Fire and Explosion

The present contribution attempts to list and discuss some of the main steps that need to be taken sequentially to guard against the risk of fire and explosion in industrial fuel installations. It also sets up the steps to be followed for combating and fighting a fire once it develops. These steps are the following:-

i) Avoid any unrequired or uncontrolled presence of fuels such as through fuel spills, combustible gas and vapour discharges, leaks, etc. Remove combustible materials, residue, rubbish, etc.

ii) Keep any fuel separated from the air or other oxidants.

iii) Keep the size of any fuel and air that may become mixed down to an absolute minimum.

iv) Keep any resulting fuel and air mixtures well outside the corresponding lean and rich flammability limits.

v) Search for and remove any likely sources for ignition of these mixtures.

vi) Restrict and confine any fire spread that may develop as far as possible.

vii) Separate and isolate the combustion region once fire takes place.

viii) Cool the burning fuel.

ix) Reduce and shut off heat transfer to the reacting system.

x) Slow down the reaction rate of the combustion system through the addition of auxiliary materials such as diluents, etc.

I. Avoid Any Unrequired or Uncontrolled Presence of Fuels

Obviously, an essential ingredient for the development of a fire is the presence of a sufficient quantity of a combustible material that may undergo, under the right conditions, exothermic chemical reaction. For example, releases of flammable gases and vapours from leaky or ruptured pipes, hoses, containers, various parts of equipment, etc., are common contributors to the onset of fire and explosion in industrial settings. Hence, every effort must be made throughout to avoid and guard against the inadvertent release of fuels, however small, from pipes, containers, reservoirs, etc. through leaks, discharges, spills, etc. Moreover, intermittent, batch or starting and shutdown operations, in contrast to continuous operations, tend to represent an increased likelihood for the undersirable accumulation of fuel, sometimes to unsafe levels. Accordingly, adequate controls must be furnished such that they provide throughout prompt shutdown or starting, without the uncontrolled accumulation of any fuel within working spaces, ducting, environment, etc.

A practice of paramount importance that must be followed at all times for preventing and guarding against fires is the prompt removal of combustible products, waste, rubbish, residue, etc. and not permitting their improper accumulation. Moreover, great care needs to be taken in ensuring throughout the proper storage of various combustibles and fuels. For example, some fuel gases, such as ethylene or acetylene, have positive heats of formation and can decompose exothermically and explosively at high pressures, even in the absence of air in the presence of adequate heat sources, igniters or catalysts, unless sufficient amount of appropriate inert diluent is mixed with the fuel.

II. Keep Any Fuel Entirely Separated from the Air or Other Oxidants

Fuel-air mixtures support combustion readily within certain concentration limits once an ignition source is provided. Arresting propagating flames within premixed fuel-air media tends to be no easy task and the energy release rate can be both very significant and rapid. However, when a quantity of the fuel in its unmixed state becomes exposed to the air, the rate of burning and the resulting rate of energy release will be controlled by the rate of mixing of the fuel and the surrounding air. Preventing or reducing the free entrainment of air above the fuel surface will stop or delay the ignition processes and slow down subsequent combustion.

Accordingly, it is imperative for safety that any fuel vapour or gas that may be released in a closed or restricted environment or the atmosphere following leakage be prevented from mixing with the air required for its combustion. Moreover, keep in mind that there are a range of chemicals that can act as effective and virulent oxidants, although they may not contain oxygen and these must be equally guarded against.

III. Keep the Size of Fuel and Air Mixtures Down to the Absolute Minimum

The total amount of energy released and the associated potential for the resulting damage will depend on the amount of fuel and air mixture that may become available for combustion. Hence, the size of the fuel and air mixture that may result from an inadvertent or controlled fuel leakage or release should be kept at all times down to the absolute minimum. Mixing of the fuel and air can be rapid when pressure, density or temperature gradients are present. A still environment produces smaller rates of mixing in contrast to flow or turbulent conditions. The presence of obstacles or moving or rotating machinery tends to promote increased mixing. Normally, the odds for fire and explosion tend to increase with large equipment and installations that would involve greater quantitative emission of flammable materials and thus result in larger quantities of fuel-air mixtures. Moreover, designs and controls should be fail safe, shutting down equipment promptly following the detection of a malfunction or a failure. Also, repair procedures must not contribute to the release of fuel and contribute to the hazard.

IV. Keep any Resulting Fuel and Air Mixtures to Well Outside the Corresponding Lean and Rich Flammability Limits

The flammability limit is a convenient criterion that has been widely used in the assessment of the potential for fire and explosion hazards arising from the release or seepage of a fuel vapour or gas into air, whether continuously or intermittently within a reservoir, a pipeline in the open atmosphere, etc. The limit would indicate for any mixture of fuel and oxidant and set of operating conditions the limiting concentrations for a flame to propagate within the homogeneous mixture following ignition from a sufficient source of energy. These limits, being the lower and higher limits of flammability cannot be predicted theoretically and relate only to a homogeneous stationary mixture under well specified conditions. For example, the influence of important common operating parameters such as mixture temperature, pressure, movement and turbulence intensity needs to be established for any fuel-oxidant system experimentally.

The lean or lower limit for many of the common organic liquid fuels in air tends to be roughly around 55% of their stoichiometric concentration in air, while for many compounds, the rich or upper limit is very approximately

3.5 times the stoichiometric concentration in air (Table I). For many of the other common gaseous fuels, these limits vary widely, depending on the type of fuel being considered. Similarly, organic combustible compounds in air in the presence of an inert gas will not support combustion if the oxygen volumetric concentration in the mixture is below roughly about 10.5% and 13.0% by volume for nitrogen and carbon dioxide respectively (6), Figure 2. Values involving water vapour inerting tend to be somewhat between those for nitrogen and carbon dioxide (7). However, these are very gross approximations and throughout reference to appropriate experimental data about the flammability limits of the mixture being considered needs to be made. The limits may vary according to the size of the containing vessel (7) and direction of flame propagation. Larger size vessels and upward resulting flame propagation widens the limits. Of course, the limits, especially the rich limit, become wider in oxygenated air. Moreover, the limits of fuel mixtures generally can be predicted from a knowledge of the corresponding limits of the components making up the mixture, particularly in relation to the lean limit (8).

Accordingly, to reduce the hazards arising from the release of a fuel vapour or gas into air, the following measures can be taken:

i) Supplying additional air to lean mixtures.

ii) Excluding additional air or adding further fuels to rich mixtures.

iii) Supplying to the mixture adequate amounts of inerts or diluents such as nitrogen, carbon dioxide, helium, steam, etc.

iv) Avoiding further heating of the mixture or resorting to its chilling.

v) Regulating process variables that influence the value of the limits, e.g. temperature, air flow, agitation, orientation to potential ignition sources, etc. so as to remain outside the flammability zone.

V. Search for and Remove any Likely Ignition Sources

To effect ignition of a combustible mixture, a minimum amount of energy is needed to be supplied locally to the mixture. The amount of this energy will depend on the fuel-air mixture and numerous other factors such as concentrations, temperature, pressure, etc. Stoichiometric mixtures are easiest to ignite. Hydrogen in contrast to methane requires merely a small amount of energy for ignition, rendering mixtures of hydrogen in air very hazardous. In practice, there is almost always the likelihood of ignition sources becoming available. Accordingly, it must not be presumed that the excluding of obvious ignition sources is an adequate procedure for ensuring safety. Nevertheless, every attempt should be made to exclude their incidence.

It is essential that equipment, electrical wiring, etc., installed shall not be capable of producing sufficient electrical or thermal energy, whether under normal or abnormal conditions, to bring about the ignition of a specific hazardous atmospheric mixture in its most easily ignitable concentration. Abnormal conditions can include accidental damage to wiring, failure of components, over voltage, adjustments and maintenance operations, overloading, etc. (9).

Throughout, the potential danger of electrostatic discharges generated from sources such as accidental releases of flammable materials, rubbing movement of parts, flow of fluids through pipes, etc., must be guarded against such as by adequate grounding (10). Moreover, guarding against ignition by sparks resulting from friction or impact should also be ensured. Similarly, great care should be exercised that uncontrolled ignition will never take place from repair procedures such as the using of tools, cutting and welding operations. Furthermore, catalytic materials such as some metals or metal oxides can enhance the oxidation process at their surfaces to such an extent, leading to high local metal surface temperature, that they can lead ultimately to the ignition of the whole mixture.

VI. Confine and Restrict Fire Spread as far as Possible

Flame propagation rates are fastest around the stoichiometric region. They are increased very significantly with increased turbulence and the presence of obstacles that may be in the path of the flame. The principle of sectioning needs to be applied to installations handling flammable materials. Moreover, the compartmentalization of certain parts can also permit different strategies and the placement of fire extinguishing systems to be applied appropriately. The individual units and sections of a plant must not only be designed to contain a limited fire, such as through the provision of fire walls, but also the connections between the units must be of similar resistance and strength.

Finally, it may be necessary to allow the fire to take its full course. It is then essential to ensure that it does so limitedly, controllably and relatively safely, with the minimum of damage. Pressure relieving facilities, whether through a relief valve or bursting diaphragms invariably may be needed. The consequences of the resulting discharge from such devices must be dealt with appropriately and safely. Dyking of fuel spills or leading spills in appropriate troughs for fighting needs also to be considered.

VII. Separate the Combustion Region Once Fire Takes Place

It is necessary to separate promptly the burning region from other parts of the combustible material once fire takes place and to apply various possible procedures to deprive the fire of its main sustaining ingredients, fuel and air. These procedures for fire suppression must be available and applied promptly.

The shutting off of the air supply to the fire is necessary through covering of openings and vents to the burning compartment or shutting doors tightly. Similarly, blanketing of fuel surfaces with foam or sand is an effective approach for most fuel fires after effecting prompt shut off procedures of fuel supplies. Drenching of the fire and its adjacent space with an inert gas such as carbon dioxide, nitrogen or halon or by direct application of dry chemicals are also very effective. Inerting techniques, however, may not be readily applicable to compartments that are either large or semi-closed since the application and subsequent loss of the inerting gas is both expensive and environmentally unacceptable. Of course, the control of a fire such as by the reduction of oxygen or inerting can pose severe life hazard to both occupants and fire fighters. Moreover, the sudden access of air following a fire into a compartment can lead to a renewed flaring or even further explosions.

VIII. Cool the Burning Fuel

There is a need to cool the burning fuel surface to below the temperature level that will produce vapours that feed the fire and continue the combustion. This type of cooling is effective when high flash point fuels are involved. However, it is less effective when low flash point fuels are involved. Water is especially effective with some solids, such as wood or paper, and with high flash point hydrocarbon liquid fuel fires, provided the water is introduced at the surface as a high velocity spray which causes droplets penetration and cooling of the surface layers. If this is not done effectively, then the water in liquid fuel fires will sink to the bottom where it may only eventually displace burning liquid fuel from its containment (11).

Air agitation sometimes when conducted appropriately may be helpful in extinguishing fires by rolling some cold liquid away from the surface to replace the heated layer near the surface.

Facilities for water spray of the exterior of fuel tanks and associated piping need to be provided to maintain the fuel temperature low, particular attention being given also to provide adequate thermal insulation to such tanks.

IX. Reduce or Shut Off Heat Transfer to the Reacting System

The spread of flame and fire within a combustible medium is only possible if the thermal energy liberated within the combustion zone is allowed to be transferred to the fresh layers of the mixture raising their temperature and propagating the reaction and the flame to fresh regions. Obviously, to arrest the propagating flame, this transport of heat and associated active species must be retarded so as to bring about a halt to the exothermic reactions. Premixed flames are extinguished by direct physical quenching where the reaction zone is cooled below a critical value which is the principal mechanism for flame arrestors. Diffusion flames can also be extinguished by cutting off the supply of fuel vapours such as through blanketing the surface with a suitable foam. Water application, which is more commonly used to cool the fuel surface, is particularly effective if introduced into the flame in the form of a fine spray, mist or as steam.

Flame arrestors normally consist of a multitude of narrow channels, each with an effective internal diameter less than the corresponding quenching distance through which the flame cannot propagate. They are effective so long as they can conduct the heat from the flame rapidly enough. Prolonged exposure to the hot flame even sometimes intermittently may render them ineffective. Flame arrestors tend to impose pressure drop and need to be maintained free and clear from clogging.

Diffusion flames may also be extinguished by the mechanism of "blow out," such as encountered when blowing out small flames and candles. The reaction zone within the flame is distorted so as to reduce its thickness and deny the fuel vapours enough time to react. If the reaction zone is too thin, then combustion will be incomplete, and the flame is effectively cooled to a level where it cannot be sustained (11).

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table I *　*TYPICAL COMBUSTION PROPERTIES OF SOME HYDROCARBON GASES AND HYDROGEN IN AIR*

Fuel	*Molecular weight*	A *Flammability limits % v/v gas Lower*	*Upper*	*% Gas at stoichiometric ratio*	B *% Gas at maximum burning velocity*	B *Maximum laminar burning velocity S_u (m/s)*	C *Adiabatic flame temperature T_f K*	*Expansion ratio T_f/T_i (T_i = 288K)*	D *Maximum laminar flame speed (m/s)*	E *Auto-ignition temperature K*	F *Minimum ignition energy, millijoules*	G *Net calorific value (288K, 1 atmos) MJ/m^3*	*Fuel*
Hydrogen H_2	2	4	75	30	54	3 5	2318	8 0	28 0	847	0 02	10 2	Hydrogen H_2
Methane CH_4	16	5	15	9 5	10	0 45	2148	7 4	3 5	813	0 29	34	Methane CH_4
Ethane C_2H_6	30	3	12 5	5 6	6 3	0 53	2168	7 5	4 0	788	0 24	60 5	Ethane C_2H_6
Propane C_3H_8	44	2 2	9 5	4 0	4 5	0 52	2198	7 6	4 0	723	0 25	86 4	Propane C_3H_8
Butane C_4H_{10}	58	1 9	8 5	3 1	3 5	0 50	2168	7 5	3 7	678	0 25	112 4	Butane C_4H_{10}
Pentane C_5H_{12}	72	1 5	7 8	2 6	2 9	0 52	2232	7 7	4 0	533	0 25	138 1	Pentane C_5H_{12}
Hexane C_6H_{14}	86	1 2	7 5	2 2	2 5	0 52	2221	7 7	4 0	498	0 25	164 4	Hexane C_6H_{14}
Heptane C_7H_{16}	100	1 2	6 7	1 9	2 3	0 52	2196	7 6	4 0	488	0 25	190 4	Heptane C_7H_{16}
Acetylene C_2H_2	26	2 5	80	7 7	9 3	1 58	2598	9 0	14 2	578	0 02	51	Acetylene C_2H_2
Ethylene C_2H_4	28	3 1	32	6 5	7 4	0 83	2248	7 8	6 5	763	0 12	56	Ethylene C_2H_4
Propylene C_3H_6	42	2 4	10 3	4 4	5 0	0 66	2208	7 7	5 1	733	0 28	81 5	Propylene C_3H_6
Butylene C_4H_8	56	1 7	9 5	3 4	3 9	0 57	2203	7 6	4 3	658	0 28	107 1	Butylene C_4H_8
Benzene C_6H_6	78	1 4	7 1	2 7	3 3	0 62	2287	7 9	4 9	833	0 22	134	Benzene C_6H_6
Cyclohexane C_6H_{12}	84	1 3	8 0	2 3	2 7	0 52	2232	7 8	4 1	518	0 24	167.3	Cyclohexane C_6H_{12}

* Ref. 5.

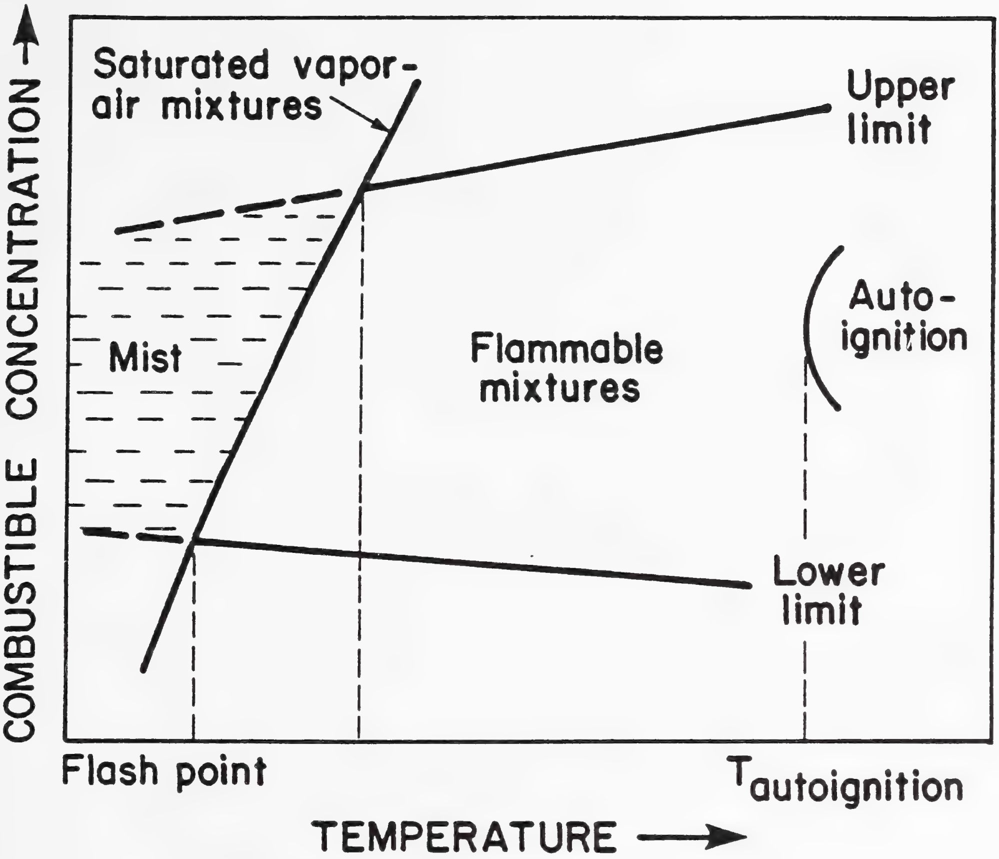

Figure 1. Schematic representation of the various regions for flammability as a function of temperature for a typical fuel-air system, including regions where fuel mist formation is considered. (Ref. 4)

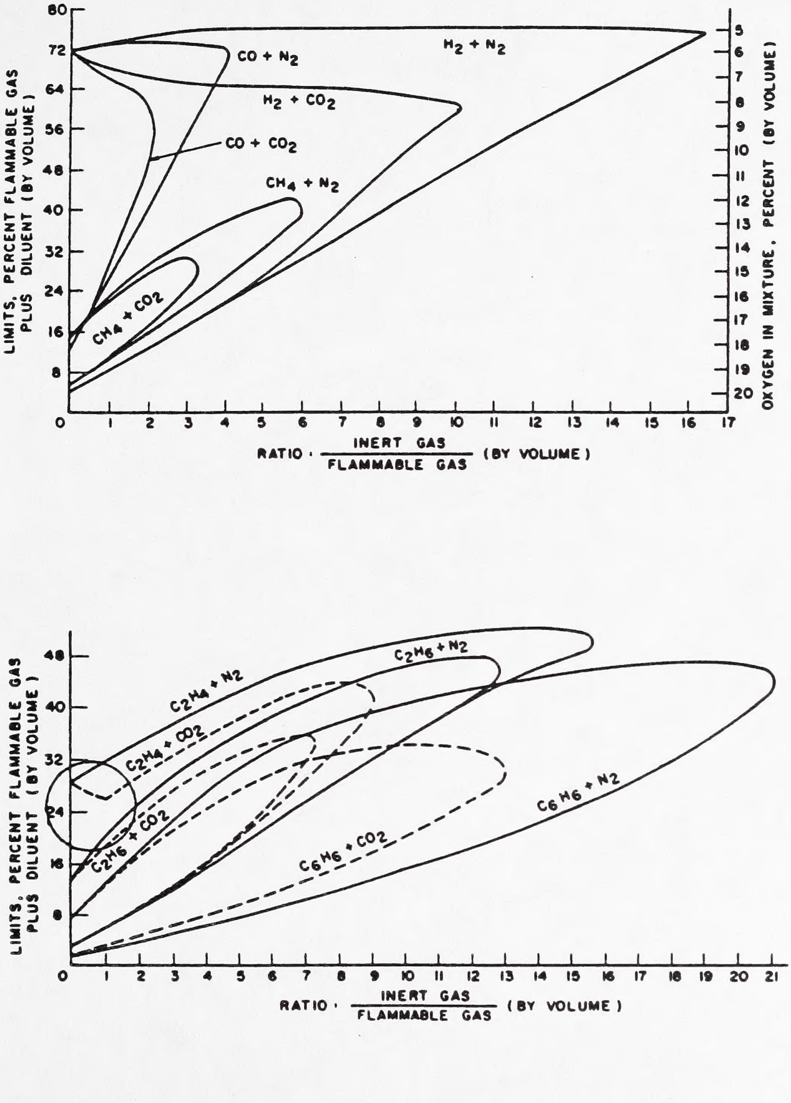

Figure 2. Typical variations of the limits of the flammability of a range of fuels with the extent of presence of the diluents N_2 and CO_2 with the fuel at ambient temperature and pressure (7).

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